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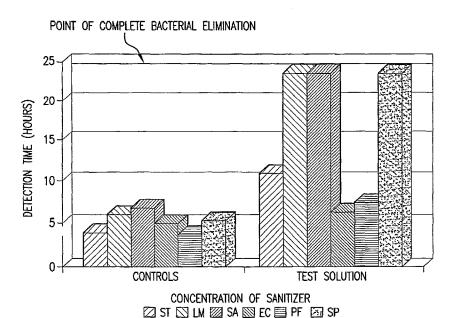
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(54) Title: COMPOSITIONS AND METHODS FOR REDUCING OR PREVENTING MICROORGANISM GROWTH OR SURVIVAL IN AQUEOUS ENVIRONMENTS



24 HOUR DETECTION TIME MEANS NO GROWTH OCCURRED AFTER EXPOSURE TO SANITIZER

(57) Abstract: Disclosed are antimicrobial compositions and methods for using such compositions to reduce, prevent, or eliminate a microorganism in aqueous environments such as recreational and industrial waters.



# COMPOSITIONS AND METHODS FOR REDUCING OR PREVENTING MICROORGANISM GROWTH OR SURVIVAL IN AQUEOUS ENVIRONMENTS

#### CROSS-REFERENCE TO RELATED APPLICATIONS

This application claims benefit of U.S. Provisional application no. 60/830,078, filed July 11, 2006, which is hereby incorporated herein by reference in its entirety.

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#### **FIELD**

The disclosed matter generally relates to compositions and methods for reducing or preventing microorganism growth or survival in aqueous environments such as recreational, industrial, agricultural, and irrigation waters.

#### BACKGROUND

Microorganism growth in aqueous environments can be a significant problem. For example, swimming pools and water parks can be ideal environments for bacterial, fungal, and algal growth because they are often around neutral pH, at ambient temperatures or above, and are contaminated on a regular basis from the environment and from patrons. The result of microorganism growth in such recreational waters can lead to serious health risks. Thus, it has been estimated that over 1 billion dollars are spent every year on services and compositions for maintaining good water quality in private, municipal, and commercial pool and spa facilities.

In other examples, microorganism growth can be problematic for industrial waters, such as cooling towers, mining process waters, food processing waters, papermaking waters, oil field waters, sugar reprocessing waters, carpet manufacturing waters, etc. Taking the paper and paperboard industry as an example, paper fibers are suspended in a large volume of recirculating water before being removed by filtration and compacted and dried to form the paper sheet. Various water-based additives are also used to improve the efficiency of the process and control the quality of the finished paper. These carbohydrate rich conditions, together with the operating temperatures, which are normally above 30°C, provide ideal environments for the growth of microorganisms.

In the paper manufacturing process, contamination can result in the build up of bacterial slime at key points in the process. If uncontrolled, the slime can accumulate and eventually break away, leading to problems such as paper breaks, spray nozzle blockage,

and discoloration of the finished product. Problem organisms include the aerobic slime forming bacteria such as *Pseudomonas*, *Klebsiella*, *Enterobacter*, and *Bacillus* spp.

Anaerobic organisms such as *Desulphovibrio* spp can also accumulate under slime layers and other deposits leading to the production of foul odors and corrosive byproducts. Also bacterial spores can impact packaging products, particularly those products used for food and dairy packaging.

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Water based mill additives including mineral slurries and paper coatings are also prone to contamination and require adequate preservation to ensure that their stability is not compromised. The major spoilage organisms are aerobic bacteria such as those mentioned above but in some instances yeasts such as *Candida* and *Saccharomyes* spp can be involved. Consequently, there can be a need for broad spectrum preservation in many of these additive products. Preservation of bulk pulp can also be required during unplanned shut-downs.

The effects of microorganism growth illustrated above for the papermaking industry are similar to the effects observed in other industrial water systems. Waters used in, for example, mining, carpet, aqua culture, and food industries are often rich in nutrients and at acceptable temperature and pH for microbial growth. Generally, microbial contamination results in the formation of slime and deposits on machines, which can corrode parts and lead to increased down time, loss of yields, odors, and costs associated with removing such deposits or replacing corroded parts.

Microbial growth can also be a problem in agricultural and irrigation waters. In such circumstances, irrigation hoses, pipes and sprayers and drip lines are used to deliver water to plants and crops. Often, such water is from natural sources, like wells and reservoirs, and contains algal, fungal, and bacterial microorganisms. Such microbes can build up inside the pipes and lines causing blockages. Moreover, nutrients and fertilizers are often added to such waters, which results in increased microbial growth in irrigation apparatus.

The conventional method for controlling microorganism grown in such waters is through the use of biocides. While biocides can inhibit microorganism growth, economic and environmental concerns require improved methods. One problem with biocides is that high levels or expensive chemicals are needed to control microbial growth. Thus, the use of many biocides requires continuous or frequent treatment. Moreover, many biocides are highly toxic in large quantities. As a result, environmental regulations restrict the amount that can be safely discarded into the environment.

Thus, there is a need for antimicrobial compositions and methods for treating recreational industrial, agricultural, and irrigation aqueous environments to remove a broad range of microorganisms. The antimicrobial compositions and methods disclosed herein meet these and other needs.

5 SUMMARY

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In accordance with the purposes of the disclosed materials, compounds, compositions, and methods, as embodied and broadly described herein, the disclosed matter, in one aspect, relates to compositions and methods for preparing and using such compositions. In another aspect, disclosed herein are antimicrobial compositions and methods for using such compositions to reduce, prevent, or eliminate one or more microorganisms in an aqueous environment.

The advantages described below will be realized and attained by means of the elements and combinations particularly pointed out in the appended claims. It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory only and are not restrictive.

#### BRIEF DESCRIPTION OF THE FIGURES

The accompanying figures, which are incorporated in and constitute a part of this specification, illustrate several aspects described below.

In the following figures "ST" refers to Salmonella typhimurium, "LM" refers to Listeria monocytogenes, "SA" refers to Staphylococcus aureau, "EC" refers to Escherichia coli, "PF" refers to Pseudomonas fluorscens, and "SP" refers to Shewanella putrefaciens. Also, an antimicrobial composition as disclosed herein is indicated as "Test Solution" and a control solution is indicated as "Controls."

Figure 1 is a graph showing the effect of an antimicrobial composition as disclosed herein and a control solution on pathogenic and spoilage bacterial isolates. The results are shown in terms of detection times (hours). Detection times of 24 hours mean no growth occurred after exposure to test solution.

Figure 2 is a graph showing the reduction of the indicated bacterial colonies (in log<sub>10</sub> colony forming units) when exposed to an antimicrobial composition as disclosed herein or a control solution.

Figure 3 is a graph showing the effect of an antimicrobial composition as disclosed herein and control solution on the indicated bacterial isolates. The results are shown in terms of detection times (hours). Detection times of 24 hours mean no growth occurred after exposure to test solution.

Figure 4 is a graph showing the reduction of indicated bacterial colonies (in log<sub>10</sub> colony forming units) when exposed to an antimicrobial composition as disclosed herein or a control solution.

Figure 5 is a graph showing the effect of an antimicrobial composition as disclosed herein and a control solution when used to treat the indicated microorganisms attached to food contact surfaces. The results are shown in terms of detection times (hours). Detection times of 24 hours mean no growth occurred after exposure to test solution.

# **DETAILED DESCRIPTION**

The materials, compositions, articles, devices, and methods described herein may be understood more readily by reference to the following detailed description of specific aspects of the disclosed subject matter, and methods and the Examples included therein and to the Figures and their previous and following description.

Before the present materials, compositions, articles, devices, and methods are disclosed and described, it is to be understood that the aspects described below are not limited to specific synthetic methods or specific reagents, as such may, of course, vary. It is also to be understood that the terminology used herein is for the purpose of describing particular aspects only and is not intended to be limiting.

Also, throughout this specification, various publications are referenced. The disclosures of these publications in their entireties are hereby incorporated by reference into this application in order to more fully describe the state of the art to which the disclosed subject matter pertains. The references disclosed are also individually and specifically incorporated by reference herein for the material contained in them that is discussed in the sentence in which the reference is relied upon.

# General Definitions

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In this specification and in the claims that follow, reference will be made to a number of terms, which shall be defined to have the following meanings:

As used in the specification and the appended claims, the singular forms "a," "an," and "the" include plural referents unless the context clearly dictates otherwise. Thus, for example, reference to "a compound" includes mixtures of two or more such compounds, reference to "an agent" includes mixtures of two or more such agents, reference to "the composition" includes mixtures of two or more such compositions, and the like.

Throughout the specification and claims, the word "comprise" and variations of the word, such as "comprising" and "comprises," means "including but not limited to,"

and is not intended to exclude, for example, other additives, components, integers, or steps.

"Optional" or "optionally" means that the subsequently described event or circumstance can or cannot occur, and that the description includes instances where the event or circumstance occurs and instances where it does not.

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Ranges can be expressed herein as from "about" one particular value, and/or to "about" another particular value. When such a range is expressed, another embodiment includes from the one particular value and/or to the other particular value. Similarly, when values are expressed as approximations, by use of the antecedent "about," it will be understood that the particular value forms another embodiment. It will be further understood that the endpoints of each of the ranges are significant both in relation to the other endpoint, and independently of the other endpoint. It is also understood that there are a number of values disclosed herein, and that each value is also herein disclosed as "about" that particular value in addition to the value itself. For example, if the value "10" is disclosed, then "about 10" is also disclosed. It is also understood that when a value is disclosed that "less than or equal to" the value, "greater than or equal to the value" and possible ranges between values are also disclosed, as appropriately understood by the skilled artisan. For example, if the value "10" is disclosed then "less than or equal to 10" as well as "greater than or equal to 10" is also disclosed. It is also understood that the throughout the application, data is provided in a number of different formats, and that this data, represents endpoints and starting points, and ranges for any combination of the data points. For example, if a particular data point "10" and a particular data point "15" are disclosed, it is understood that greater than, greater than or equal to, less than, less than or equal to, and equal to 10 and 15 are considered disclosed as well as between 10 and 15. It is also understood that each unit between two particular units are also disclosed. For example, if 10 and 15 are disclosed, then 11, 12, 13, and 14 are also disclosed.

References in the specification and claims to parts by weight of a particular element or component in a composition or article denotes the weight relationship between the element or component and any other elements or components in the composition or article for which a part by weight is expressed. Thus, in a compound containing 2 parts by weight of component X and 5 parts by weight component Y, X and Y are present at a weight ratio of 2:5, and are present in such ratio regardless of whether additional components are contained in the compound.

A weight percent of a component, unless specifically stated to the contrary, is

based on the total weight of the formulation or composition in which the component is included.

By "reduce" or other forms of the word, such as "reducing" or "reduction," is meant lowering of an event or characteristic (e.g., microorganism growth or survival). It is understood that this is typically in relation to some standard or expected value, in other words it is relative, but that it is not always necessary for the standard or relative value to be referred to. For example, "reduces the population of bacteria" means lowering the amount of bacteria relative to a standard or a control.

By "prevent" or other forms of the word, such as "preventing" or "prevention," is meant to stop a particular event or characteristic, to stabilize or delay the development or progression of a particular event or characteristic, or to minimize the chances that a particular event or characteristic will occur. Prevent does not require comparison to a control as it is typically more absolute than, for example, reduce. As used herein, something could be reduced but not prevented, but something that is reduced could also be prevented. Likewise, something could be prevented but not reduced, but something that is prevented could also be reduced. It is understood that where reduce or prevent are used, unless specifically indicated otherwise, the use of the other word is also expressly disclosed.

By "treat" or other forms of the word, such as "treated" or "treatment," is meant to administer a composition or to perform a method in order to reduce, prevent, inhibit, break-down, or eliminate a particular characteristic or event (e.g., microorganism growth or survival).

By "antimicrobial" is meant the ability to treat (e.g., reduce, prevent, inhibit, break-down, or eliminate) microorganism growth or survival at any concentration.

# Chemical Definitions

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As used herein, the term "substituted" is contemplated to include all permissible substituents of organic compounds. In a broad aspect, the permissible substituents include acyclic and cyclic, branched and unbranched, carbocyclic and heterocyclic, and aromatic and nonaromatic substituents of organic compounds. Illustrative substituents include, for example, those described below. The permissible substituents can be one or more and the same or different for appropriate organic compounds. For purposes of this disclosure, the heteroatoms, such as nitrogen and oxygen, can have hydrogen substituents and/or any permissible substituents of organic compounds described herein which satisfy the valences of the heteroatoms. This disclosure is not intended to be limited in any

manner by the permissible substituents of organic compounds. Also, the terms "substitution" or "substituted with" include the implicit proviso that such substitution is in accordance with permitted valence of the substituted atom and the substituent, and that the substitution results in a stable compound, e.g., a compound that does not spontaneously undergo transformation such as by rearrangement, cyclization, elimination, etc. Also, as used herein "substitution" or "substituted with" is meant to encompass configurations where one substituent is fused to another substituent. For example, an aryl group substituted with an aryl group (or vice versa) can mean that one aryl group is bonded to the second aryl group via a single sigma bond and also that the two aryl groups are fused, e.g., two carbons of one alkyl group are shared with two carbons of the other aryl group.

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"A<sup>1</sup>," "A<sup>2</sup>," "A<sup>3</sup>," and "A<sup>4</sup>" are used herein as generic symbols to represent various specific substituents. These symbols can be any substituent, not limited to those disclosed herein, and when they are defined to be certain substituents in one sentence it does not mean that, in another sentence, they cannot be defined as some other substituents.

The term "alkyl" as used herein is a branched or unbranched saturated hydrocarbon group of 1 to 40 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, t-butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, dodecyl, tetradecyl, hexadecyl, octadecyl, eicosyl, tetracosyl, and the like. The alkyl group can also be substituted or unsubstituted. The alkyl group can be substituted with one or more groups including, but not limited to, alkyl, halogenated alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfooxo, sulfonylamino, nitro, silyl, or thiol, as described below.

Throughout the specification "alkyl" is generally used to refer to both unsubstituted alkyl groups and substituted alkyl groups; however, substituted alkyl groups are also specifically referred to herein by identifying the specific substituent(s) on the alkyl group. For example, the term "alkyl halide" specifically refers to an alkyl group that is substituted with one or more halides, e.g., fluorine, chlorine, bromine, or iodine. When "alkyl" is used in one sentence and a specific term such as "alkyl halide" is used in another, it is not meant to imply that the term "alkyl" does not also refer to specific terms such as "alkyl halide" and the like.

This practice is also used for other groups described herein. That is, while a term such as "heteroaryl" refers to both unsubstituted and substituted heteroaryl moieties, the

substituted moieties can, in addition, be specifically identified herein; for example, a particular substituted heteroaryl can be referred to as, e.g., an "alkyl heteroaryl." Similarly, a substituted alkenyl can be, e.g., an "alkenyl halide," and the like. Again, the practice of using a general term, such as "heteroaryl," and a specific term, such as "alkyl heteroaryl," is not meant to imply that the general term does not also include the specific term.

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The term "alkenyl" as used herein is a hydrocarbon group of from 2 to 40 carbon atoms with a structural formula containing at least one carbon-carbon double bond. Asymmetric structures such as  $(A^1A^2)C=C(A^3A^4)$  are intended to include both the E and Z isomers. This may be presumed in structural formulae herein wherein an asymmetric alkene is present, or it may be explicitly indicated by the bond symbol C=C. The alkenyl group can be substituted with one or more groups including, but not limited to, alkyl, halogenated alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfo-oxo, sulfonylamino, nitro, silyl, or thiol.

The term "alkynyl" as used herein is a hydrocarbon group of 2 to 40 carbon atoms with a structural formula containing at least one carbon-carbon triple bond. The alkynyl group can be substituted with one or more groups including, but not limited to, alkyl, halogenated alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfo-oxo, sulfonylamino, nitro, silyl, or thiol.

The term "aliphatic" as used herein refers to a non-aromatic hydrocarbon group and includes branched and unbranched alkyl, alkenyl, or alkynyl groups.

The term "aryl" as used herein is a group that contains any carbon-based aromatic group including, but not limited to, benzene, benzyl, naphthalene, phenyl, biphenyl, phenoxybenzene, and the like. The term "aryl" also includes "heteroaryl," which is defined as a group that contains an aromatic group that has at least one heteroatom incorporated within the ring of the aromatic group. Examples of heteroatoms include, but are not limited to, nitrogen, oxygen, sulfur, and phosphorus. Likewise, the term "non-heteroaryl," which is also included in the term "aryl," defines a group that contains an aromatic group that does not contain a heteroatom. The aryl group can be substituted or unsubstituted. The aryl group can be substituted with one or more groups including, but not limited to, alkyl, halogenated alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfo-oxo, sulfonylamino, or thiol as described herein. The term "biaryl" is a specific type of aryl group and is included in the definition of aryl. Biaryl refers to two aryl groups that are

bound together via a fused ring structure, as in naphthalene, or are attached via one or more carbon-carbon bonds, as in biphenyl.

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The term "cycloalkyl" as used herein is a non-aromatic carbon-based ring composed of at least three carbon atoms. Examples of cycloalkyl groups include, but are not limited to, cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc. The term "heterocycloalkyl" is a cycloalkyl group as defined above where at least one of the carbon atoms of the ring is substituted with a heteroatom such as, but not limited to, nitrogen, oxygen, sulfur, or phosphorus. The cycloalkyl group and heterocycloalkyl group can be substituted or unsubstituted. The cycloalkyl group and heterocycloalkyl group can be substituted with one or more groups including, but not limited to, alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfo-oxo, sulfonylamino, nitro, silyl, or thiol.

The term "cycloalkenyl" as used herein is a non-aromatic carbon-based ring composed of at least three carbon atoms and contains at least one double bound, e.g., C=C. Examples of cycloalkenyl groups include, but are not limited to, cyclopropenyl, cyclobutenyl, cyclopentenyl, cyclopentadienyl, cyclohexenyl, cyclohexadienyl, and the like. The term "heterocycloalkenyl" is a type of cycloalkenyl group as defined above, and is included within the meaning of the term "cycloalkenyl," where at least one of the carbon atoms of the ring is substituted with a heteroatom such as, but not limited to, nitrogen, oxygen, sulfur, or phosphorus. The cycloalkenyl group and heterocycloalkenyl group can be substituted or unsubstituted. The cycloalkenyl group and heterocycloalkenyl group can be substituted with one or more groups including, but not limited to, alkyl, alkoxy, alkenyl, alkynyl, aryl, heteroaryl, aldehyde, amino, carboxylic acid, ester, ether, halide, hydroxy, ketone, sulfo-oxo, sulfonylamino, nitro, silyl, or thiol.

The term "cyclic group" is used herein to refer to either aryl groups (e.g., heteraryl, biaryl), non-aryl groups (i.e., cycloalkyl, heterocycloalkyl, cycloalkenyl, and heterocycloalkenyl groups), or both. Cyclic groups have one or more ring systems that can be substituted or unsubstituted. A cyclic group can contain one or more aryl groups, one or more non-aryl groups, or one or more aryl groups and one or more non-aryl groups.

The terms "amine" or "amino" as used herein are represented by the formula:

$$A^1$$
 $N \longrightarrow A^2$ 

where A<sup>1</sup>, A<sup>2</sup>, and A<sup>3</sup> can each be, independent of one another, hydrogen, an alkyl, halogenated alkyl, alkenyl, alkynyl, aryl, heteroaryl, cycloalkyl, cycloalkenyl, heterocycloalkyl, or heterocycloalkenyl group described above. Also, any of the A<sup>1</sup>, A<sup>2</sup>, and A<sup>3</sup> substituents can be absent and any of the remaining substituents can be a multivalent group, *i.e.*, form more than one bond with N.

The terms "ammonium" or "quaternary ammonium" are represented by the formula:

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$$A^{4} \xrightarrow{A^{1}} A^{2}$$

$$A^{3}$$

where A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup>, and A<sup>4</sup> can each be, independent of one another, hydrogen, an alkyl, halogenated alkyl, alkenyl, alkynyl, aryl, heteroaryl, cycloalkyl, cycloalkenyl, heterocycloalkyl, or heterocycloalkenyl group described above. Also, any of the A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup>, and A<sup>4</sup> substituents can be absent and any of the remaining substituents can be a multivalent group.

The term "halide" as used herein refers to the halogens fluorine, chlorine, bromine, and iodine.

"X," "R<sup>1</sup>," "R<sup>2</sup>," and "R<sup>n</sup>," where n is some integer, as used herein can, independently, possess two or more of the groups listed above. For example, if R is a straight chain alkyl group, one of the hydrogen atoms of the alkyl group can optionally be substituted with a hydroxyl group (OH), an alkoxy group, halide, etc. Depending upon the groups that are selected, a first group can be incorporated within second group or, alternatively, the first group can be pendant (*i.e.*, attached) or fused to the second group.

Unless stated to the contrary, a formula with chemical bonds shown only as solid lines and not as wedges or dashed lines contemplates each possible isomer, e.g., each enantiomer and diastereomer, and a mixture of isomers, such as a racemic or scalemic mixtures.

Reference will now be made in detail to specific aspects of the disclosed materials, compounds, compositions, components, devices, articles, and methods,

examples of which are illustrated in the following description and examples, and in the figures and their previous and following description.

# Methods

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Disclosed herein are methods of treating one or more microorganisms in an aqueous environment that comprises contacting the aqueous environment with an effective amount of an antimicrobial composition. By the term "effective amount" of a composition as provided herein is meant an amount of a composition sufficient to provide the desired result, e.g., reduction or prevention of microorganism growth or survival. As will be pointed out below, the exact amount required will vary from use to use, depending on the type of aqueous environment, the type of microorganism to be treated, the size of the processing facilities (e.g., the volume of the water), the mode of application, the particular compositions being used, and the like. Thus, it is not possible to specify an exact "effective amount." However, an appropriate effective amount can be determined by one of ordinary skill in the art using only routine experimentation.

While it is not possible to specify an exact amount, the disclosed antimicrobial 15 compositions can be used neat or diluted in a ratio as described herein. Also, when diluted to form an aqueous solution, an amount of the disclosed antimicrobial compositions can be used such that an aqueous environment will contain about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 20 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120, 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 154, 25 155, 156, 157, 158, 159, 160, 161, 162, 163, 164, 165, 166, 167, 168, 169, 170, 171, 172, 173, 174, 175, 176, 177, 178, 179, 180, 181, 182, 183, 184, 185, 186, 187, 188, 189, 190, 191, 192, 193, 194, 195, 196, 197, 198, 199, 200, 201, 202, 203, 204, 205, 206, 207, 208, 209, 210, 211, 212, 213, 214, 215, 216, 217, 218, 219, 220, 221, 222, 223, 224, 225, 226, 227, 228, 229, 230, 231, 232, 233, 234, 235, 236, 237, 238, 239, 240, 241, 242, 243, 244, 30 245, 246, 247, 248, 249, 250, 251, 252, 253, 254, 255, 256, 257, 258, 259, 260, 261, 262, 263, 264, 265, 266, 267, 268, 269, 270, 271, 272, 273, 274, 275, 276, 277, 278, 279, 280, 281, 282, 283, 284, 285, 286, 287, 288, 289, 290, 291, 292, 293, 294, 295, 296, 297, 298. 299, 300, 301, 302, 303, 304, 305, 306, 307, 308, 309, 310, 311, 312, 313, 314, 315, 316,

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As compared with other treatment methods, the disclosed compositions and methods have been found particularly advantageous in that the treatment process is faster and less caustic. In addition, because a smaller amount of the disclosed antibacterial compositions is used, the process is more effective. Also, the disclosed antimicrobial compositions, in most cases, do not require complex equipment for their application, removal, recycling, or disposal. The following non-limiting examples, further illustrate advantages of the disclosed compositions and methods over other antimicrobial solutions and processes.

For example, the disclosed methods can result in a plate count at 35°C of not more than 200 colonies per 10 mL. When treating coliform, the disclosed methods can result in less than 2.2 organisms per 100 mL or no more than 2 *enterococcal* organisms per 50 mL.

# Treatable Microorganisms

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As disclosed herein, the antimicrobial compositions can be used to treat various

surfaces to reduce, inhibit, prevent, disrupt, degrade, brake-down, eliminate, etc. microorganism growth or survival. By "microorganism" or "microbe" is meant a single or multicelled organism, and can include one or more organisms of the same type or mixtures of organism. The microorganisms that can be treated by the compositions and methods disclosed herein can be Gram-positive or Gram-negative bacteria. Such bacteria can be pathogenic, indicator, and/or spoilage bacteria. In one aspect, the antimicrobial compositions disclosed herein can be used to treat microorganisms in aqueous environments.

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The Gram-positive bacteria treatable by the compositions and methods disclosed herein can include, but are not limited to, M. tuberculosis, M. bovis, M. typhimurium, M. bovis strain BCG, BCG substrains, M. avium, M. intracellulare, M. africanum, M. kansasii, M. marinum, M. ulcerans, M. avium subspecies paratuberculosis, Staphylococcus aureus, Staphylococcus epidermidis, Staphylococcus equi, Streptococcus pyogenes, Streptococcus agalactiae, Listeria monocytogenes, Listeria ivanovii, Bacillus anthracis, B. subtilis, Nocardia asteroides, and other Nocardia species, Streptococcus viridans group, Peptococcus species, Peptostreptococcus species, Actinomyces israelii and other Actinomyces species, Propionibacterium acnes, and Enterococcus species.

The Gram-negative bacteria treatable by the compositions and methods disclosed herein can include, but are not limited to, Clostridium tetani, Clostridium perfringens, Clostridium botulinum, other Clostridium species, Pseudomonas aeruginosa, other 20 Pseudomonas species, Campylobacter species, Vibrio cholerae, Ehrlichia species, Actinobacillus pleuropneumoniae, Pasteurella haemolytica, Pasteurella multocida, other Pasteurella species, Legionella pneumophila, other Legionella species, Salmonella typhi, other Salmonella species, Shigella species Brucella abortus, other Brucella species, Chlamydi trachomatis, Chlamydia psittaci, Coxiella burnetti, Escherichia coli, Neiserria 25 meningitidis, Neiserria gonorrhea, Haemophilus influenzae, Haemophilus ducreyi, other Hemophilus species, Yersinia pestis, Yersinia enterolitica, other Yersinia species, Escherichia coli, Escherichia hirae and other Escherichia species, as well as other Enterobacteriacae, Brucella abortus and other Brucella species, Burkholderia cepacia, Burkholderia pseudomallei, Francisella tularensis, Bacteroides fragilis, Fusobascterium 30 nucleatum, Provetella species, Cowdria ruminantium, Klebsiella species, and Proteus species.

The above examples of Gram-positive, Gram-negative, pathogenic, indicator, and spoilage bacteria are not intended to be limiting, but are intended to be representative of a

larger population including all bacteria that effect public health, as well as non-Gram test responsive bacteria. Examples of other species of microorganisms include, but are not limited to, Abiotrophia, Achromobacter, Acidaminococcus, Acidovorax, Acinetobacter, Actinobacillus, Actinobaculum, Actinomadura, Actinomyces, Aerococcus, Aeromonas, Afipia, Agrobacterium, Alcaligenes, Alloiococcus, Alteromonas, Amycolata, 5 Amycolatopsis, Anaerobospirillum, Anaerorhabdus, Arachnia, Arcanobacterium, Arcobacter, Arthrobacter, Atopobium, Aureobacterium, Bacteroides, Balneatrix, Bartonella, Bergeyella, Bifidobacterium, Bilophila Branhamella, Borrelia, Bordetella, Brachyspira, Brevibacillus, Brevibacterium, Brevundimonas, Brucella, Burkholderia, Buttiauxella, Butyrivibrio, Calymmatobacterium, Campylobacter, Capnocytophaga, 10 Cardiobacterium, Catonella, Cedecea, Cellulomonas, Centipeda, Chlamydia, Chlamydophila. Chromobacterium, Chyseobacterium, Chryseomonas, Citrobacter, Clostridium, Collinsella, Comamonas, Corynebacterium, Coxiella, Cryptobacterium, Delftia, Dermabacter, Dermatophilus, Desulfomonas, Desulfovibrio, Dialister, Dichelobacter, Dolosicoccus, Dolosigranulum, Edwardsiella, Eggerthella, Ehrlichia, 15 Eikenella, Empedobacter, Enterobacter, Enterococcus, Erwinia, Erysipelothrix, Escherichia, Eubacterium, Ewingella, Exiguobacterium, Facklamia, Filifactor, Flavimonas, Flavobacterium, Francisella, Fusobacterium, Gardnerella, Globicatella, Gemella, Gordona, Haemophilus, Hafnia, Helicobacter, Helococcus, Holdemania Ignavigranum, Johnsonella, Kingella, Klebsiella, Kocuria, Koserella, Kurthia, 20 Kytococcus, Lactobacillus, Lactococcus, Lautropia, Leclercia, Legionella, Leminorella, Leptospira, Leptotrichia, Leuconostoc, Listeria, Listonella, Megasphaera, Methylobacterium, Microbacterium, Micrococcus, Mitsuokella, Mobiluncus, Moellerella, Moraxella, Morganella, Mycobacterium, Mycoplasma, Myroides, Neisseria, Nocardia, Nocardiopsis, Ochrobactrum, Oeskovia, Oligella, Orientia, Paenibacillus, Pantoea, 25 Parachlamydia, Pasteurella, Pediococcus, Peptococcus, Peptostreptococcus, Photobacterium, Photorhabdus, Plesiomonas, Porphyrimonas, Prevotella, Propionibacterium, Proteus, Providencia, Pseudomonas, Pseudonocardia, Pseudoramibacter, Psychrobacter, Rahnella, Ralstonia, Rhodococcus, Rickettsia Rochalimaea, Roseomonas, Rothia, Ruminococcus, Salmonella, Selenomonas, Serpulina, 30 Serratia, Shewenella, Shigella, Simkania, Slackia, Sphingobacterium, Sphingomonas, Spirillum, Staphylococcus, Stenotrophomonas, Stomatococcus, Streptobacillus, Streptococcus, Streptomyces, Succinivibrio, Sutterella, Suttonella, Tatumella, Tissierella. Trabulsiella, Treponema, Tropheryma, Tsakamurella, Turicella, Ureaplasma,

Vagococcus, Veillonella, Vibrio, Weeksella, Wolinella, Xanthomonas, Xenorhabdus, Yersinia, and Yokenella.

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In specific examples, the disclosed antimicrobial compositions can be used to treat the bacteria *Pseudomonas aeruginosa*, *Enterobacter aerogenes*, *Proteus vulgaris*, *Staphylococcus aureus*, *Bacillus cereus*, *Escherichia coli*, and *Legionella pneumophila*. Further, sulfur reducing bacteria can be problematic in oil field waters, and even in drinking water. Such sulfur reducing bacteria can be treated by the compositions and methods disclosed herein. Some specific, non-limiting examples of sulfur reducing bacteria that can be treated by the disclosed compositions and methods are species of the genera *Desulfovibrio*, *Desulfotomaculum*, *Desulfomonas*, *Desulfobulbus*, *Desulfobacter*, *Desulfococcus*, *Desulfonema*, *Desulfosarcina*, *Desulfobacterium*, and *Desulforomas*.

The disclosed antimicrobial compositions can be used to treat other microorganisms such as, for example, parasites. Examples of parasites that can be treated include, but are not limited to, *Toxoplasma gondii*, *Plasmodium* species such as *Plasmodium falciparum*, *Plasmodium vivax*, *Plasmodium malariae*, and other *Plasmodium* species, *Trypanosoma brucei*, *Trypanosoma cruzi*, *Leishmania* species such as *Leishmania major*, *Schistosoma* such as *Schistosoma mansoni* and other *Shistosoma* species, and *Entamoeba histolytica*.

The disclosed antimicrobial compositions can also be used to treat fungal species such as, but not limited to, Candida albicans, Cryptococcus neoformans, Histoplama capsulatum, Aspergillus fumigatus, Coccidiodes immitis, Paracoccidiodes brasiliensis, Blastomyces dermitidis, Pneomocystis carnii, Penicillium marneffi, Alternaria alternata, and Fusarium species. In other examples, the microorganism that can be treated can comprise the fungi Aspergilium niger, Aspergilius phoenicis, Penicillium funiculosum, Alternaria alternata, Cladosporium cladosporioides, Endomyces geotrichum, Aerobasidium pullulan, or Chaetomium globosum.

The disclosed antimicrobial compositions can also be used to treat algal species such as, but not limited to, Oscillatoria geminate, Nostoc sp, Phormidium foveolarum, Chlorella vulgaris, Chlorella pyrenoidosa, Scenedesmus sp, Ulthrix subtilissima, and Tribonema aequale.

In one particularly useful example, the disclosed compositions and methods can be used to treat aqueous environments that contain avian flu virus. Such virus can be present in water bowls, ponds, animal habitats, or in standing water. Consumption or contact with such contaminated waters can lead to infection with the avian virus. By

using the compositions as disclosed herein, infection can be prevented. The disclosed compositions can also be used to destroy avian flu virus in blood sera.

# **Aqueous Environments**

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The disclosed methods can be used to treat microbial contamination in a variety of aqueous environments. In general, any aqueous system that one desires to prevent and/or reduce microbial growth and survival can be treated by the disclosed methods. While the disclosed methods are not limited by the particular aqueous environment, suitable examples include recreational industrial, agricultural, and irrigation waters. Examples of recreational aqueous environments contemplated herein are swimming pools, hot tubes, spas, Jacuzzi's, water parks (e.g., slides and flumes), and the like. Also contemplated are fountains, decorative ponds, fish ponds (e.g., Koi ponds, goldfish ponds), water features, and the like, which are commonly used in indoor and outdoor landscaping. Further examples of aqueous environments include, but are not limited to, animal habitat waters, such as would be found in a zoo.

Examples of industrial aqueous environments contemplated herein are cooling waters (e.g., water used in cooling towers), oil field waters (e.g., water used in wells), mining process waters, food processing waters, papermaking waters, sugar reprocessing waters, carpet manufacturing waters, and the like. Thus, water used in any industrial process which may be subject to microbial contamination can be treated by the methods and compositions disclosed herein.

Examples of agricultural and irrigation waters are reservoirs, wells, irrigation lines, hoses, aqua ducts, sprayers, sprinklers, drip lines, and soaker hoses. Also, the disclosed compositions can be used to treat various fungi that contaminate irrigation waters by contacting such fungi with the disclosed compositions.

In a specific example, the disclosed compositions can be used in biocidal applications to control bacteria growth, biofilm growth, and sulphate reducing bacteria, or SRB in water systems used for drilling and processing various hydrocarbon resources. Bacteria control in such oil field waters can reduce the amount of "sour" gas and oil by limiting the amount of bacteria-produced sulphur compounds and control of sulphate reducing bacteria can reduce the amount of microbiological induced corrosion (MIC) of metal components.

In further examples, the aqueous environment can be aqueous culture waters.

Many marine species are cultured in aqueous environments for a source of food and other desired natural products. And oftentimes, the aqueous culture waters can become

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contaminated with undesirable microbes, which can be treated as disclosed herein. Examples of marine species that are cultured in aqueous environments, which can be treated by the compositions and methods disclosed herein, include, but are not limited to, shrimp, bivalve mollusks, e.g., oysters and clams, herbivorous zooplankton, such as brine shrimp, (Artemia salina) and fish. Exemplary fish include, but are not limited to, Oncorhynchus kisutch (coho salmon), Paralichthys lethostigma (southern flounder), and Hippoglossus hippglossus (Atlantic halibut). Aqueous culture waters for the shrimp Litopenaeus vannamei, Penaeus japonicus, Penaeus orientalis, and Penaeus monodon, and the shellfish Crassostrea gigas (Pacific oyster), Ostrea edulis (flat oyster), Patinopecten vessoensis (deep sea scallop), Argopecten irradians (bay scallop), and Haliotis rufescens (red abalone) can also be treated by the methods disclosed herein. The list above is merely an abbreviated list of the marine animals that can be cultured in the aqueous culture waters treated with the compositions disclosed herein. Further, microalgae are also cultured in aqueous culture waters and can be treated by the disclosed compositions. Examples of suitable microalgae are set forth in the CRC Handbook of Microalgal Mass Culture (ed. Amos Richmond, Boca Raton, Fla.: CRC Press, c1986), which is incorporated by reference herein for its teachings of types of microalgae. Other suitable marine algae are all those included in the Culture Collections of Algae worldwide, some of which include: 1) American Type Culture Collection (ATCC), P.O. Box 1549, Manassas, Va. 20108, USA; 2) Culture Collection of Algae and Protozoa (CCAP), Scottish Association for Marine Science, Dunstaffnage Marine Laboratory, Dunbeg, OBAN, Argyll PA37 1QA, United Kingdom; 3) Canadian Center for the Culture of Microorganisms, The North East Pacific Culture Collection (NEPCC), Department of Botany, 6270 University Boulevard, Vancouver, B.C. Canada V6T 1Z4; 4) Centro de Investigaciones Biologicas del Noroeste (CIBNOR), S.C., Mar Bermejo No. 195, Col. Playa Palo de Santa Rita, Apdo. Postal 128; La Paz, BCS 23090, Mexico; 5) CSIRO Marine Research (CS), GdPO Box 1538, Hobart, Tasmania, 7001, Australia; 6) Provasoli-Guillard National Center for Culture of Marine Phytoplankton (CCMP), McKown Point, West Boothbay Harbor, Me. 04575; 7) UTEX Culture Collection of Algae/MCDB (UTEX), 1 University Station A6700, The University of Texas at Austin, 30 Austin, Tex. 78712-0183, USA. Aqueous culture waters can be found in natural and manmade ponds, lakes, tanks, bioreactors, hatcheries, and the like.

In many examples, suitable aqueous waters have a pH of from about 4 to about 9, from about 5 to about 8, or from about 6 to about 7. Also, suitable aqueous environments

can be at a temperature of from about 4°C to about 45°C, from about 10°C to about 30°C, or at ambient temperature. Also, many suitable aqueous environments that can be treated by the methods and compositions disclosed herein contain cellulosic material, carbohydrates, and/or chlorine or bromine.

In still another example, the aqueous environment can be drinking water for an animal (including human drinking water). Such waters include pre-treated waters, such as those in a reservoir, but which are to be used for drinking.

# Compositions

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Disclosed herein, in one aspect, are antimicrobial compositions. The disclosed antimicrobial compositions can be used to treat various aqueous environments in order to eliminate, reduce, and/or prevent microorganism growth or survival.

The materials and components that can be used for, can be used in conjunction with, can be used in preparation for, or are products of the disclosed compositions and methods are disclosed herein, and it is understood that when combinations, subsets, interactions, groups, etc. of these materials are disclosed that while specific reference of each various individual and collective combinations and permutation of these compounds may not be explicitly disclosed, each is specifically contemplated and described herein. For example, if a molecule is disclosed and a number of modifications that can be made to a number of substituents are discussed, each and every combination and permutation that are possible are specifically contemplated unless specifically indicated to the contrary. In another example, if a composition is disclosed and a number of modifications that can be made to a number of components in the composition are discussed, each and every combination and permutation that are possible are specifically contemplated unless specifically indicated to the contrary. Thus, if a class of substituents or components A, B, and C are disclosed as well as a class of substituents or components D, E, and F and an example of a combination molecule or combination composition, A-D is disclosed, then even if each is not individually recited, each is individually and collectively contemplated. Thus, in this example, each of the combinations A-E, A-F, B-D, B-E, B-F, C-D, C-E, and C-F are specifically contemplated and should be considered disclosed from disclosure of A, B, and C; D, E, and F; and the example combination A-D. Likewise, any subset or combination of these is also specifically contemplated and disclosed. Thus, for example, the sub-group of A-E, B-F, and C-E are specifically contemplated and should be considered disclosed from disclosure of A, B, and C; D, E, and F; and the example combination A-D. This concept applies to all aspects of this

disclosure including, but not limited to, steps in methods of making and using the disclosed compositions. Thus, if there are a variety of additional steps that can be performed it is understood that each of these additional steps can be performed with any specific embodiment or combination of embodiments of the disclosed methods, and that each such combination is specifically contemplated and should be considered disclosed.

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The disclosed antimicrobial compositions, in one aspect, comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, and at least two ammonium salts comprising an aliphatic heteroaryl salt, a dialiphatic dialkyl ammonium salt, or a tetraalkyl ammonium salt. For example, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, an aliphatic heteroaryl salt, and a tetraalkyl ammonium salt. In another example, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt. In a still further example, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, an aliphatic heteroaryl ammonium salt, and a dialiphatic dialkyl ammonium salt.

In a further aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt and trichloromelamine. The disclosed antimicrobial compositions can also comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, and an aliphatic heteroaryl ammonium salt (e.g., alkyl pyridinium halide, trichloromelamine, and alkyl benzylalkyl ammonium halide). In yet a further aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, and a dialiphatic dialkyl ammonium salt. In a still further aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, and a tetraalkyl ammonium salt. In a further aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, an aliphatic heteroaryl ammonium salt, and a tetraalkyl ammonium salt. In yet a further aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt.

In a preferred aspect, the disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt (e.g., alkyldimethylbenzalkonium halide) and trichloromelamine.

# Aliphatic heteroaryl salt

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The disclosed antimicrobial compositions can comprise an aliphatic heteroaryl salt (e.g., one or more aliphatic heteroaryl salts). An aliphatic heteroaryl salt is a compound that comprises an aliphatic moiety bonded to a heteroaryl moiety, and a counterion, as are defined herein. One or more types of aliphatic heteroaryl salts can be used in the antimicrobial compositions disclosed herein.

# Aliphatic moiety

In the aliphatic heteroaryl salt component of the disclosed antimicrobial compositions, the aliphatic moiety can be any alkyl, alkenyl, alkynyl, cycloalkyl, or cycloalkenyl group, as described herein. Generally, the aliphatic moiety can comprise at least 10, at least 12, at least 14, at least 16, at least 18, or at least 20 carbon atoms. In other examples, the aliphatic moiety can comprise a mixture of aliphatic groups having a range of carbon atoms. For example, the aliphatic moiety can comprise from 10 to 40, from 12 to 38, from 14 to 36, from 16 to 34, from 18 to 32, from 14 to 18, or from 20 to 30 carbon atoms. In some specific examples, the aliphatic moiety can contain 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 carbon atoms, where any of the stated values can form an upper or lower endpoint when appropriate. Examples of specific aliphatic moieties that can be used in the disclosed aliphatic heteroaryl salts include, but are not limited to, decyl, dodecyl (lauryl), tetradecyl (myristyl), hexadecyl (palmityl or cetyl), octadecyl (stearyl), eicosyl (arachidyl), and linolenyl groups, including branched derivatives thereof and any mixtures thereof. In the aliphatic heteroaryl salts, the aliphatic moiety is bonded to a heteroatom in the heteroaryl moiety.

# Heteroaryl moiety

In the aliphatic heteroaryl salt component of the disclosed antimicrobial compositions, the heteroaryl moiety can be any heteroaryl moiety as described herein. For example, the heteroaryl moiety can be an aryl group having one or more heteroatoms. Examples of specific heteroaryl moieties that can be used in the aliphatic heteroaryl salts include, but are not limited to, pyrazole, pyridine, pyrazine, pyrimidine, pryidazine, indolizine, isoindole, indole, indazole, imidazole, oxazole, triazole., thiazole, purine, isoquinoline, quinoline, phthalazine, quinooxaline, phenazine, and the like, including substituted derivatives and mixtures thereof.

In the aliphatic heteroaryl salts, a heteroatom in the heteroaryl moiety is bonded to the aliphatic moiety. When the heteroatom is nitrogen, this forms a quaternary

ammonium species.

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#### Counterion

In the disclosed aliphatic heteroaryl salts, the counterion can be any ion that has an opposite charge as the remaining aliphatic heteroaryl portion of the salt. For example, when the heteroatom of heteroaryl moiety is bonded to the aliphatic moiety to form a positively charged quaternary ammonium moiety, the counterion can be a negatively charged moiety. Likewise, if the aliphatic heteroaryl portion is negatively charged, then the counterion can be positively charged. In the disclosed aliphatic heteroaryl salts, one or more different types of counterions can be present.

In some specific examples, the counterion can be a halide, such as a fluoride, chloride, bromide, or iodide. In other examples, suitable counterions for the aliphatic heteroaryl salt can include, but are not limited to, sulfide, sulfates, sulfites, phosphide, phosphates, phosphites, carbonates, bicarbonates, nitrates, nitrites, hypochlorite, chlorite, perchlorate, acetate, formate, hydroxide, and the like, including mixtures thereof.

# Specific Examples

In one aspect, the aliphatic heteroaryl salt can have any of the aliphatic moieties disclosed above combined with any of the heteroaryl moieties disclosed above. In some specific examples, the aliphatic heteroaryl salt can be an alkyl pyridinium salt, an alkyl quinolinium salt, an alkyl imidazolinium salt, or any mixture thereof. In other examples, the aliphatic heteroaryl salt can be an alkenyl pyrazolium salt, an alkenyl pyrazinium salt, an alkenyl quinolinium salt, or any mixture thereof. The counter ions for these specific examples can be halides, nitrates, sulfates, carbonates or any other counterion disclosed herein. In other aspects, a specific example of an alkyl pyridinium salt includes an alkyl pyridinium halide such as, but not limited to, cetylpyridinium halide (e.g., cetylpyridinium chloride, cetylpyridinium bromide, or mixtures thereof), laurylpyridinium halide (e.g., laurylpyridinium chloride, laurylpyridinium bromide, or mixtures thereof), myristylpyridinium halide (e.g., myristylpyridinium chloride, myristylpyridinium bromide, or mixtures thereof), stearylpyridinium halide (e.g., stearylpyridinium chloride, stearylpyridinium bromide, or mixtures thereof), and arachidylpyridinium halide (arachidylpyridinium chloride, arachidylpyridinium bromide, or mixtures thereof). In a specific example, the aliphatic heteroaryl salt can comprise cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof.

#### **Amounts**

The aliphatic heteroaryl salts disclosed herein can be prepared by methods known

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in the art or can be obtained from commercial sources. The aliphatic heteroaryl salt can be present in the antimicrobial compositions disclosed herein in an amount of from less than about 20 weight %, less than about 15 weight %, less than about 10 weight %, less than about 8 weight %, less than about 6 weight %, less than about 5 weight %, less than about 4 weight %, less than about 3 weight %, less than about 2 weight %, less than about 1 weight %, or less than about 0.5 weight %, based on the total weight of the antimicrobial composition. In another aspect, the aliphatic heteroaryl salt can be present in the antimicrobial compositions disclosed herein in an amount of from greater than about 0.5 weight %, greater than about 1 weight %, greater than about 2 weight %, greater than about 3 weight %, greater than about 4 weight %, greater than about 5 weight %, greater than about 6 weight %, greater than about 8 weight %, greater than about 10 weight %, greater than about 15 weight %, or greater than about 20 weight %, based on the total weight of the antimicrobial composition. In still another aspect, the aliphatic heteroaryl salt can be present in the antimicrobial compositions disclosed herein in an amount of from about 0.5 to about 20 weight %, from about 1 to about 15 weight %, from about 2 to about 10 weight %, from about 3 to about 8 weight %, from about 3.5 to about 8 weight %, from about 4 to about 6 weight %, from about 6 to about 8 weight %, or about 7.5 weight %, based on the total weight of the antimicrobial composition. In yet another aspect, the aliphatic heteroaryl salt can be present in the antimicrobial compositions disclosed herein in an amount of about 0.25, 0.5, 0.75, 1.0, 1.25, 1.5, 1.75, 2.0, 2.25, 2.5, 2.75, 3.0, 3.25, 3.5, 3.75, 4.0, 4.25, 4.5, 4.75, 5.0, 5.25, 5.5, 5.75, 6.0, 6.25, 6.5, 6.75, 7.0, 7.25, 7.5, 7.75, 8.0, 8.25, 8.5, 8.75, 9.0, 9.25, 9.5, 9.75, 10.0, 10.25. 10.5, 10.75, 11.0, 11.25, 11.5, 11.75, 12.0, 12.25, 12.5, 12.75, 13.0, 13.25, 13.5, 13.75, 14.0, 14.25, 14.5, 14.75, 15.0, 15.25, 15.5, 15.75, 16.0, 16.25, 16.5, 16.75, 17.0, 17.25, 17.5, 17.75, 18.0, 18.25, 18.5, 18.75, 19.0, 19.25, 19.5, 19.75, or 20.0 weight %, based on the total weight of the antimicrobial composition and where any of the stated values can form an upper or lower endpoint when appropriate.

In another aspect, the antimicrobial compositions disclosed herein can contain less than about 20 parts by weight, less than about 15 parts by weight, less than about 10 parts by weight, less than about 8 parts by weight, less than about 6 parts by weight, less than about 5 parts by weight, less than about 4 parts by weight, less than about 3 parts by weight, less than about 2 parts by weight, less than about 1 part by weight, or less than about 0.5 parts by weight of the aliphatic heteroaryl salt. In another aspect, the antimicrobial compositions disclosed herein can contain greater than about 0.5 parts by

weight, greater than about 1 part by weight, greater than about 2 parts by weight, greater than about 3 parts by weight, greater than about 4 parts by weight, greater than about 5 parts by weight, greater than about 6 parts by weight, greater than about 8 parts by weight, greater than about 10 parts by weight, greater than about 15 parts by weight, or greater than about 20 parts by weight of the aliphatic heteroaryl salt. In still another aspect, the antimicrobial compositions disclosed herein can contain from about 0.5 to about 20 parts by weight, from about 1 to about 15 parts by weight, from about 2 to about 10 parts by weight, from about 3 to about 8 parts by weight, from about 3.5 to about 8 parts by weight, from about 4 to about 6 parts by weight, from about 6 to about 8 parts by weight, or about 7.5 parts by weight of the aliphatic heteroaryl salt. In yet another aspect, the antimicrobial compositions disclosed herein can contain about 0.25, 0.5, 0.75, 1.0, 1.25, 1.5, 1.75, 2.0, 2.25, 2.5, 2.75, 3.0, 3.25, 3.5, 3.75, 4.0, 4.25, 4.5, 4.75, 5.0, 5.25, 5.5, 5.75, 6.0, 6.25, 6.5, 6.75, 7.0, 7.25, 7.5, 7.75, 8.0, 8.25, 8.5, 8.75, 9.0, 9.25, 9.5, 9.75, 10.0, 10.25, 10.5, 10.75, 11.0, 11.25, 11.5, 11.75, 12.0, 12.25, 12.5, 12.75, 13.0, 13.25, 13.5, 13.75, 14.0, 14.25, 14.5, 14.75, 15.0, 15.25, 15.5, 15.75, 16.0, 16.25, 16.5, 16.75, 17.0, 17.25, 17.5, 17.75, 18.0, 18.25, 18.5, 18.75, 19.0, 19.25, 19.5, 19.75, or 20.0 parts by weight of the aliphatic heteroaryl salt, where any of the stated values can form an upper or lower endpoint when appropriate.

# Trichloromelamine

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The disclosed antimicrobial compositions comprise trichloromelamine. Trichloromelamine (i.e., N²,N⁴,N⁶-Trichloro-2,4,6-triamino-s-triazine) can be prepared by methods known in the art or can be obtained from commercial sources. Trichloromelamine can be present in the antimicrobial compositions disclosed herein in any amount as is described above for the aliphatic benzylalkyl ammonium salt. For example, trichloromelamine can be present in an amount of from in an amount of from less than about 1.0 weight %, less than about 0.75 weight %, less than about 0.5 weight %, less than about 0.25 weight %, less than about 0.0075 weight %, less than about 0.005 weight %, less than about 0.001 weight %, based on the total weight of the antimicrobial composition. In another aspect, trichloromelamine can be present in the antimicrobial compositions disclosed herein in an amount of from greater than about 0.001 weight %, greater than about 0.005 weight %, greater than about 0.005 weight %, greater than about 0.005 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.001 weight %, greater than about 0.0075 weight %, greater than about 0.0075 weight %, greater than about 0.0075 weight %.

0.025 weight %, greater than about 0.05 weight %, greater than about 0.075 weight %, greater than about 0.1 weight %, greater than about 0.25 weight %, greater than about 0.5 weight %, greater than about 0.75 weight %, or greater than about 1.0 weight %, based on the total weight of the antimicrobial composition. In still another aspect, trichloromelamine can be present in the antimicrobial compositions disclosed herein in an

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amount of from about 0.001 to about 1.0 weight %, from about 0.0025 to about 0.75 weight %, from about 0.005 to about 0.5 weight %, 0.005 to about 0.1 weight %, from about 0.0075 to about 0.25 weight %, from about 0.01 to about 0.1 weight %, from about 0.025 to about 0.075 weight %, about 0.005 to about 0.1 weight %, about 0.005 to about 0.02 weight %, about 0.005 to about 0.01 weight %, or about 0.01 weight %, based on the total weight of the antimicrobial composition. Still further, trichloromelamine can be present in an amount of from about 0.001 to about 0.1 weight %, from about 0.005 to about 0.075 weight %, from about 0.0075 about 0.05 weight %, or from about 0.01 to about 0.02 weight %, based on the total weight of the antimicrobial composition. In yet another aspect, trichloromelamine can be present in the antimicrobial compositions disclosed herein in an amount of about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009,0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 weight %, based on the total weight of the antimicrobial composition and where any of the stated values can form an upper or lower endpoint when appropriate.

In another examples, the disclosed antimicrobial compositions can contain less than about 1.0 parts by weight, less than about 0.75 parts by weight, less than about 0.5 parts by weight, less than about 0.05 parts by weight, less than about 0.075 parts by weight, less than about 0.05 parts by weight, less than about 0.05 parts by weight, less than about 0.025 parts by weight, less than about 0.0075 parts by weight, less than about 0.0075 parts by weight, less than about 0.0025 parts by weight, or less than about 0.001 parts by weight of trichloromelamine. In another aspect, the antimicrobial compositions disclosed herein can contain greater than about 0.001 parts by weight, greater than about 0.005 parts by weight, greater than about 0.005 parts by weight, greater than about 0.01 parts by weight, greater than about 0.025 parts by weight, greater than about 0.05 parts by

weight, greater than about 0.075 parts by weight, greater than about 0.1 parts by weight, greater than about 0.25 parts by weight, greater than about 0.5 parts by weight, greater than about 0.75 parts by weight, or greater than about 1.0 parts by weight of trichloromelamine. In still another aspect, the antimicrobial compositions disclosed herein can contain from about 0.001 to about 1.0 parts by weight, from about 0.0025 to about 0.75 parts by weight, from about 0.005 to about 0.5 parts by weight, 0.005 to about 0.1 parts by weight, from about 0.0075 to about 0.25 parts by weight, from about 0.01 to about 0.1 parts by weight, from about 0.025 to about 0.075 parts by weight, about 0.005 to about 0.1 parts by weight, about 0.005 to about 0.02 parts by weight, about 0.005 to about 0.01 parts by weight, or about 0.01 parts by weight of trichloromelamine. Still further, trichloromelamine can be present in an amount of from about 0.001 to about 0.1 parts by weight, from about 0.005 to about 0.075 parts by weight, from about 0.0075 about 0.05 parts by weight, or from about 0.01 to about 0.02 parts by weight trichloromelamine. In yet another aspect, the antimicrobial compositions disclosed herein can contain about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 parts by weight of trichloromelamine, where any of the stated values can form an upper or lower endpoint when appropriate.

# Aliphatic benzylalkyl ammonium salt

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The disclosed antimicrobial compositions can comprise an aliphatic benzylalkyl ammonium salt (e.g., one or more aliphatic benzylakyl ammoniums salts). An aliphatic benzylalkyl ammonium salt is a compound that comprises an aliphatic moiety bonded to the nitrogen atom of a benzylalkyl amine moiety, and a counterion, as are defined herein. The aliphatic moiety and counterion can be as described above. The benzylalkyl amine moiety can be a benzyl amine where the amine is bonded to an alkyl or cyclic alkyl group, as described above. One or more types of aliphatic benzylalkyl ammonium salts can be used in the antimicrobial compositions disclosed herein. The aliphatic benzylalkyl ammonium salts suitable for use herein can be prepared by methods known in the art or can be obtained from commercial sources.

In one aspect, the aliphatic benzylalkyl ammonium salt can be represented by the

following formula:

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$$\begin{array}{c|c}
 & R^1 & X^1 \\
 & R^2 \\
 & R^3
\end{array}$$

wherein R<sup>1</sup> is an aliphatic group, as described above, R<sup>2</sup> and R<sup>3</sup> are, independent of one another, alkyl groups or cyclic alkyl groups as described herein, and X is a counterion as described herein. In some examples, one or more of the "R" substituents can be a long chain alkyl group (e.g., the number of carbon atoms is greater than 6). In other examples, one or more of the "R" substituents can be a short chain alkyl group (e.g., the number of carbon atoms is 6 or less). In still other examples, one of the "R" substituents is a long chain alkyl group and the other two "R" substituents are short chain alkyl groups.

Specific Examples

In one aspect, the aliphatic benzylalkyl ammonium salt can have any of the aliphatic moieties disclosed above bonded to any benzylalkyl amine moieties disclosed above. In some specific examples, R<sup>1</sup> in the formula of aliphatic benzylalkyl ammonium salts can be an aliphatic group of from 10 to 40 carbon atoms, e.g., a decyl, dodecyl (lauryl), tetradecyl (myristyl), hexadecyl (palmityl or cetyl), octadecyl (stearyl), or eicosyl (arachidyl) group, and R<sup>2</sup> and R<sup>3</sup> can each be, independent of one another, a methyl, ethyl, propyl, butyl, pentyl, or hexyl group.

In another aspect, the aliphatic benzylalkyl ammonium salts can include, but are not limited to, alkyl dimethyl benzyl ammonium halides (e.g., alkyl dimethyl benzyl ammonium chloride, alkyl dimethyl benzyl ammonium bromide, or mixtures thereof). Specific examples of alkyl dimethyl benzyl ammonium halides include, but are not limited to, cetyl dimethyl benzyl ammonium halide (e.g., cetyl dimethyl benzyl ammonium chloride, or mixtures thereof), lauryl dimethyl benzyl ammonium chloride bromide, or mixtures thereof), lauryl dimethyl benzyl ammonium bromide, or mixtures thereof), myristyl dimethyl benzyl ammonium halide (e.g., myristyl dimethyl benzyl ammonium chloride, myristyl dimethyl benzyl ammonium bromide, or mixtures thereof), stearyl dimethyl benzyl ammonium halide (e.g., stearyl dimethyl benzyl ammonium chloride, stearyl dimethyl benzyl ammonium bromide, or mixtures thereof), and arachidyl dimethyl benzyl ammonium halide (e.g., arachidyl dimethyl benzyl ammonium chloride, arachidyl dimethyl benzyl ammonium bromide, or mixtures thereof).

In yet another aspect, the aliphatic benzylalkyl ammonium salts can include, but are not limited to, alkyl methylethyl benzyl ammonium halides. Specific examples of alkyl methylethyl benzyl ammonium halides include, but are not limited to, cetyl methylethyl benzyl ammonium halide (e.g., cetyl methylethyl benzyl ammonium chloride, cetyl methylethyl benzyl ammonium chloride bromide, or mixtures thereof), lauryl methylethyl benzyl ammonium halide (e.g., lauryl methylethyl benzyl ammonium chloride, lauryl methylethyl benzyl ammonium bromide, or mixtures thereof), myristyl methylethyl benzyl ammonium halide (e.g., myristyl methylethyl benzyl ammonium chloride, myristyl methylethyl benzyl ammonium bromide, or mixtures thereof), stearyl methylethyl benzyl ammonium halide (e.g., stearyl methylethyl benzyl ammonium chloride, stearyl methylethyl benzyl ammonium bromide, or mixtures thereof), and arachidyl methylethyl benzyl ammonium halide (e.g., arachidyl methylethyl benzyl ammonium bromide, or mixtures thereof).

Aliphatic benzylalkyl ammonium salts with melamine are particularly preferred compositions.

#### Amounts

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The aliphatic benzylalkyl ammonium salts disclosed herein can be prepared by methods known in the art or can be obtained from commercial sources. The aliphatic benzylalkyl ammonium salt can be present in the disclosed antimicrobial compositions in an amount of from less than about 1.0 weight %, less than about 0.75 weight %, less than about 0.5 weight %, less than about 0.25 weight %, less than about 0.10 weight %, less than about 0.075 weight %, less than about 0.05 weight %, less than about 0.025 weight %, less than about 0.01 weight %, less than about 0.0075 weight %, less than about 0.005 weight %, less than about 0.0025 weight %, or less than about 0.001 weight %, based on the total weight of the antimicrobial composition. In another aspect, the aliphatic benzylalkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of from greater than about 0.001 weight %, greater than about 0.0025 weight %, greater than about 0.005 weight %, greater than about 0.0075 weight %, greater than about 0.01 weight %, greater than about 0.025 weight %, greater than about 0.05 weight %, greater than about 0.075 weight %, greater than about 0.1 weight %, greater than about 0.25 weight %, greater than about 0.5 weight %, greater than about 0.75 weight %, or greater than about 1.0 weight %, based on the total weight of the antimicrobial composition. In still another aspect, the aliphatic benzylalkyl ammonium

salt can be present in the antimicrobial compositions disclosed herein in an amount of from about 0.001 to about 1.0 weight %, from about 0.0025 to about 0.75 weight %, from about 0.005 to about 0.5 weight %, 0.005 to about 0.1 weight %, from about 0.0075 to about 0.25 weight %, from about 0.01 to about 0.1 weight %, from about 0.025 to about 0.075 weight %, about 0.005 to about 0.1 weight %, about 0.005 to about 0.02 weight %, about 0.005 to about 0.01 weight %, or about 0.01 weight %, based on the total weight of the antimicrobial composition. Still further, the aliphatic benzylalkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 weight %, from about 0.005 to about 0.075 weight %, from about 0.0075 about 0.05 weight %, or from about 0.01 to about 0.02 weight %, based on the total weight of the antimicrobial composition. In yet another aspect, the aliphatic benzylalkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 weight %, based on the total weight of the antimicrobial composition and where any of the stated values can form an upper or lower endpoint when appropriate.

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In another aspect, the disclosed antimicrobial compositions can contain less than about 1.0 parts by weight, less than about 0.75 parts by weight, less than about 0.5 parts by weight, less than about 0.25 parts by weight, less than about 0.10 parts by weight, less than about 0.075 parts by weight, less than about 0.005 parts by weight, less than about 0.0075 parts by weight, less than about 0.0075 parts by weight, less than about 0.0075 parts by weight, less than about 0.0025 parts by weight, or less than about 0.001 parts by weight of the aliphatic benzylalkyl ammonium salt. In another aspect, the antimicrobial compositions disclosed herein can contain greater than about 0.001 parts by weight, greater than about 0.0025 parts by weight, greater than about 0.005 parts by weight, greater than about 0.0075 parts by weight, greater than about 0.01 parts by weight, greater than about 0.025 parts by weight, greater than about 0.05 parts by weight, greater than about 0.05 parts by weight, greater than about 0.05 parts by weight, greater than about 0.10 parts by weight, greater than about 0.25 parts by weight, greater than about 0.5 parts by weight, greater than about 0.75 parts by weight, or greater than about 1.0 parts by weight of the aliphatic

benzylalkyl ammonium salt. In still another aspect, the antimicrobial compositions disclosed herein can contain from about 0.001 to about 1.0 parts by weight, from about 0.0025 to about 0.75 parts by weight, from about 0.005 to about 0.5 parts by weight, 0.005 to about 0.1 parts by weight, from about 0.0075 to about 0.25 parts by weight, from about 0.01 to about 0.1 parts by weight, from about 0.025 to about 0.075 parts by weight, about 0.005 to about 0.1 parts by weight, about 0.005 to about 0.02 parts by weight, about 0.005 to about 0.01 parts by weight, or about 0.01 parts by weight of the aliphatic benzylalkyl ammonium salt. Still further, the aliphatic benzylalkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 parts by weight, from about 0.005 to about 0.075 parts by weight, from about 0.0075 about 0.05 parts by weight, or from about 0.01 to about 0.02 parts by weight. In yet another aspect, the antimicrobial compositions disclosed herein can contain about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 parts by weight of the aliphatic benzylalkyl ammonium salt, where any of the stated values can form an upper or lower endpoint when appropriate.

# Dialiphatic dialkyl Ammonium Salts

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The disclosed antimicrobial compositions can comprise a dialiphatic dialkyl ammonium salt (e.g., one or more dialiphatic dialkyl ammonium salts). A dialiphatic dialkyl ammonium salt is a compound that comprises two aliphatic moieties and two alkyl moieties bonded to a nitrogen atom, and a counterion, as are defined herein. The aliphatic moieties can be the same or different and can be any aliphatic group as described above. The alkyl moieties can be the same or different can be any alkyl group as described above. The counterion can also be as described above. In the disclosed dialiphatic dialkyl ammoniums salts, the two aliphatic moieties can have more than 10 carbon atoms and the two alkyl moieties can have less than 10 carbon atoms. In another alternative, the two aliphatic moieties can have less than 10 carbon atoms and the two alkyl moieties can have less than 10 carbon atoms and the two alkyl moieties can have more than 10 carbon atoms. One or more types of dialiphatic dialkyl ammonium salts can be used in the antimicrobial compositions disclosed herein.

In some particular examples, the dialiphatic dialkyl ammonium salt can be didodecyl dimethyl ammonium chloride or bromide, di-tetradecyl dimethyl ammonium

chloride or bromide, dihexadecyl dimethyl ammonium chloride or bromide, and the like, including combinations thereof.

#### Amounts

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The dialiphatic dialkyl ammonium salts disclosed herein can be prepared by methods known in the art or can be obtained from commercial sources. The dialiphatic dialkyl ammonium salt can be present in the disclosed antimicrobial compositions in an amount of from less than about 1.0 weight %, less than about 0.75 weight %, less than about 0.5 weight %, less than about 0.25 weight %, less than about 0.10 weight %, less than about 0.075 weight %, less than about 0.05 weight %, less than about 0.025 weight %, less than about 0.01 weight %, less than about 0.0075 weight %, less than about 0.005 weight %, less than about 0.0025 weight %, or less than about 0.001 weight %, based on the total weight of the antimicrobial composition. In another aspect, the dialiphatic dialkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of from greater than about 0.001 weight %, greater than about 0.0025 weight %, greater than about 0.005 weight %, greater than about 0.0075 weight %, greater than about 0.01 weight %, greater than about 0.025 weight %, greater than about 0.05 weight %, greater than about 0.075 weight %, greater than about 0.1 weight %, greater than about 0.25 weight %, greater than about 0.5 weight %, greater than about 0.75 weight %, or greater than about 1.0 weight %, based on the total weight of the antimicrobial composition. In still another aspect, the dialiphatic dialkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of from about 0.001 to about 1.0 weight %, from about 0.0025 to about 0.75 weight %, from about 0.005 to about 0.5 weight %, 0.005 to about 0.1 weight %, from about 0.0075 to about 0.25 weight %, from about 0.01 to about 0.1 weight %, from about 0.025 to about 0.075 weight %, about 0.005 to about 0.1 weight %, about 0.005 to about 0.02 weight %, about 0.005 to about 0.01 weight %, or about 0.01 weight %, based on the total weight of the antimicrobial composition. Still further, the dialiphatic dialkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 weight %, from about 0.005 to about 0.075 weight %, from about 0.0075 about 0.05 weight %, or from about 0.01 to about 0.02 weight %, based on the total weight of the antimicrobial composition. In yet another aspect, the dialiphatic dialkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135,

0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 weight %, based on the total weight of the antimicrobial composition and where any of the stated values can form an upper or lower endpoint when appropriate.

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In another aspect, the disclosed antimicrobial compositions can contain less than about 1.0 parts by weight, less than about 0.75 parts by weight, less than about 0.5 parts by weight, less than about 0.25 parts by weight, less than about 0.10 parts by weight, less than about 0.075 parts by weight, less than about 0.05 parts by weight, less than about 0.025 parts by weight, less than about 0.01 parts by weight, less than about 0.0075 parts by weight, less than about 0.005 parts by weight, less than about 0.0025 parts by weight, or less than about 0.001 parts by weight of the dialiphatic dialkyl ammonium salt. In another aspect, the antimicrobial compositions disclosed herein can contain greater than about 0.001 parts by weight, greater than about 0.0025 parts by weight, greater than about 0.005 parts by weight, greater than about 0.0075 parts by weight, greater than about 0.01 parts by weight, greater than about 0.025 parts by weight, greater than about 0.05 parts by weight, greater than about 0.075 parts by weight, greater than about 0.1 parts by weight, greater than about 0.25 parts by weight, greater than about 0.5 parts by weight, greater than about 0.75 parts by weight, or greater than about 1.0 parts by weight of the dialiphatic dialkyl ammonium salt. In still another aspect, the antimicrobial compositions disclosed herein can contain from about 0.001 to about 1.0 parts by weight, from about 0.0025 to about 0.75 parts by weight, from about 0.005 to about 0.5 parts by weight, 0.005 to about 0.1 parts by weight, from about 0.0075 to about 0.25 parts by weight, from about 0.01 to about 0.1 parts by weight, from about 0.025 to about 0.075 parts by weight, about 0.005 to about 0.1 parts by weight, about 0.005 to about 0.02 parts by weight, about 0.005 to about 0.01 parts by weight, or about 0.01 parts by weight of the dialiphatic dialkyl ammonium salt. Still further, the dialiphatic dialkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 parts by weight, from about 0.005 to about 0.075 parts by weight, from about 0.0075 about 0.05 parts by weight, or from about 0.01 to about 0.02 parts by weight. In yet another aspect, the antimicrobial compositions disclosed herein can contain about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135,  $0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0\overset{\circ}{1}65, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019,$ 

0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 parts by weight of the dialiphatic dialkyl ammonium salt, where any of the stated values can form an upper or lower endpoint when appropriate.

# **Tetraalkyl Ammonium Salts**

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The disclosed antimicrobial compositions can also comprise a tetraalkyl ammonium salt (e.g., one or more tetraalkyl ammonium salts). Suitable tetraalkyl ammonium salts comprise four alkyl moieties, as disclosed herein, and a counterion, also disclosed herein. In one example, a tetralkyl ammonium salt can comprise one long chain alkyl moiety (e.g., greater than 10 carbon atoms in length) and three short chain alkyl moieties (e.g., 10 carbon atoms or less in length).

Some specific examples of tetraalkyl ammonium salts that can be included in the disclosed antimicrobial compositions include, but are not limited to, cetyl trimethyl ammonium halide (e.g., chloride or bromide), lauryl trimethyl ammonium halide (e.g., chloride or bromide), myristyl trimethyl ammonium halide (e.g., chloride or bromide), stearyl trimethyl ammonium halide (e.g., chloride or bromide), arachidyl trimethyl ammonium halide (e.g., chloride or bromide), or mixtures thereof. Other examples include, but are not limited to, cetyl dimethylethyl ammonium bromide, lauryl dimethylethyl ammonium chloride, lauryl dimethylethyl ammonium bromide, myristyl dimethylethyl ammonium chloride, myristyl dimethylethyl ammonium bromide, stearyl dimethylethyl ammonium bromide, arachidyl dimethylethyl ammonium chloride, arachidyl dimethylethyl ammonium bromide, or mixtures thereof.

#### Amounts

The tetraalkyl ammonium salts disclosed herein can be prepared by methods known in the art or can be obtained from commercial sources. The tetraalkyl ammonium salt can be present in the disclosed antimicrobial compositions in an amount of from less than about 1.0 weight %, less than about 0.75 weight %, less than about 0.5 weight %, less than about 0.25 weight %, less than about 0.10 weight %, less than about 0.075 weight %, less than about 0.025 weight %, less than about 0.025 weight %, less than about 0.005 weight %, less than about 0

than about 0.001 weight %, greater than about 0.0025 weight %, greater than about 0.005 weight %, greater than about 0.0075 weight %, greater than about 0.01 weight %, greater than about 0.025 weight %, greater than about 0.05 weight %, greater than about 0.075 weight %, greater than about 0.1 weight %, greater than about 0.25 weight %, greater than about 0.5 weight %, greater than about 0.75 weight %, or greater than about 1.0 weight %, based on the total weight of the antimicrobial composition. In still another aspect, the tetraalkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of from about 0.001 to about 1.0 weight %, from about 0.0025 to about 0.75 weight %, from about 0.005 to about 0.5 weight %, 0.005 to about 0.1 weight %, from about 0.0075 to about 0.25 weight %, from about 0.01 to about 0.1 weight %, from about 0.025 to about 0.075 weight %, about 0.005 to about 0.1 weight %, about 0.005 to about 0.02 weight %, about 0.005 to about 0.01 weight %, or about 0.01 weight %, based on the total weight of the antimicrobial composition. Still further, the tetraalkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 weight %, from about 0.005 to about 0.075 weight %, from about 0.0075 about 0.05 weight %, or from about 0.01 to about 0.02 weight %, based on the total weight of the antimicrobial composition. In yet another aspect, the tetraalkyl ammonium salt can be present in the antimicrobial compositions disclosed herein in an amount of about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075,0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 weight %, based on the total weight of the antimicrobial composition and where any of the stated values can form an upper or lower endpoint when appropriate.

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In another aspect, the disclosed antimicrobial compositions can contain less than about 1.0 parts by weight, less than about 0.75 parts by weight, less than about 0.5 parts by weight, less than about 0.10 parts by weight, less than about 0.075 parts by weight, less than about 0.075 parts by weight, less than about 0.05 parts by weight, less than about 0.005 parts by weight, less than about 0.0075 parts by weight, less than about 0.0075 parts by weight, less than about 0.0075 parts by weight, less than about 0.0025 parts by weight, or less than about 0.001 parts by weight of the tetraalkyl ammonium salt. In another aspect, the antimicrobial compositions disclosed herein can contain greater than about

0.001 parts by weight, greater than about 0.0025 parts by weight, greater than about 0.005 parts by weight, greater than about 0.0075 parts by weight, greater than about 0.01 parts by weight, greater than about 0.025 parts by weight, greater than about 0.05 parts by weight, greater than about 0.075 parts by weight, greater than about 0.1 parts by weight, greater than about 0.25 parts by weight, greater than about 0.5 parts by weight, greater than about 0.75 parts by weight, or greater than about 1.0 parts by weight of the tetraalkyl ammonium salt. In still another aspect, the antimicrobial compositions disclosed herein can contain from about 0.001 to about 1.0 parts by weight, from about 0.0025 to about 0.75 parts by weight, from about 0.005 to about 0.5 parts by weight, 0.005 to about 0.1 parts by weight, from about 0.0075 to about 0.25 parts by weight, from about 0.01 to about 0.1 parts by weight, from about 0.025 to about 0.075 parts by weight, about 0.005 to about 0.1 parts by weight, about 0.005 to about 0.02 parts by weight, about 0.005 to about 0.01 parts by weight, or about 0.01 parts by weight of the tetraalkyl ammonium salt. Still further, the tetraalkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 parts by weight, from about 0.005 to about 0.075 parts by weight, from about 0.0075 about 0.05 parts by weight, or from about 0.01 to about 0.02 parts by weight. In yet another aspect, the antimicrobial compositions disclosed herein can contain about 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035, 0.004, 0.0045, 0.005, 0.005, 0.0055, 0.006, 0.0065, 0.007, 0.0075, 0.008, 0.0085, 0.009, 0.009, 0.0095, 0.01, 0.0105, 0.011, 0.0115, 0.012, 0.0125, 0.013, 0.013, 0.0135, 0.014, 0.0145, 0.015, 0.0155, 0.016, 0.0165, 0.017, 0.017, 0.0175, 0.018, 0.0185, 0.019, 0.0195, 0.02, 0.0205, 0.021, 0.021, 0.0215, 0.022, 0.0225, 0.023, 0.0235, 0.024, 0.0245, 0.025, 0.025, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, or 1 parts by weight of the tetraalkyl ammonium salt, where any of the stated values can form an upper or lower endpoint when appropriate.

# **Additional Components**

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In addition to the components disclosed above, the disclosed antimicrobial compositions can be in the form of an aqueous solution, thus, water can be another component of the disclosed compositions. Also, the disclosed antimicrobial compositions can optionally include one or more additional components such as carriers, adjuvants, solubilizing agents, suspending agents, diluents, surfactants, other antimicrobial agents, preservatives, fillers, and additives designed to affect the viscosity, thixotropy or ability of the antimicrobial composition to adhere to and/or penetrate tissue. In one aspect, it can be desired that one or more of the additional components be consumer acceptable. By

"consumer acceptable" is meant a material that is not biologically or otherwise undesirable when consumed, e.g., an agent that is acceptable when used in or on foods and beverages and which can be consumed by an individual (e.g., human, pet, livestock, etc.) along with the selected active components without causing significant undesirable biological effects or interacting in a deleterious manner with any of the other components of the composition in which it is contained. For example, a consumer acceptable agent can be any compound generally recognized as safe (GRAS). These additional components can be prepared by methods known in the art or obtained from commercial sources.

In one example, suitable additional components include surfactants such as Triton X-100 (*i.e.*, polyethylene glycol P-1,1,3,3-tetramethylbutylphenyl ether) for better cell penetration.

#### **Carriers**

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In other examples, the antimicrobial compositions disclosed herein can further comprise a carrier. The term "carrier" means a compound, composition, substance, or structure that, when in combination with a compound or composition disclosed herein, aids or facilitates preparation, storage, administration, delivery, effectiveness, selectivity, or any other feature of the compound or composition for its intended use or purpose. For example, a carrier can be selected to minimize any degradation of the active components and to minimize any adverse side effects. Examples of suitable aqueous and non-aqueous carriers, diluents, solvents include water, ethanol, polyols (propyleneglycol, polyethyleneglycol, glycerol, and the like), vegetable oils, and suitable mixtures thereof.

#### Adjuvants

In a further example, the antimicrobial compositions disclosed herein can also comprise adjuvants such as preserving, wetting, emulsifying, suspending agents, flocculating, and dispensing agents. Prevention of the action of other microorganisms can be accomplished by various antifungal agents, for example, parabens, chlorobutanol, phenol, sorbic acid, and the like. It may also be desirable to include surfactants, binders, as for example, carboxymethylcellulose, alignates, gelatin, polyvinylpyrrolidone, sucrose, and acacia, humectants, as for example, glycerol, wetting agents, as for example, cetyl alcohol, and glycerol monostearate, adsorbents, as for example, kaolin and bentonite, and lubricants, as for example, talc, calcium stearate, magnesium stearate, solid polyethylene glycols, sodium lauryl sulfate, or mixtures thereof. Suitable flocculating agents that can be used include, but are not limited to, aluminum salts (e.g., aluminium sulphate), ferrous

salts, and ferric salts (e.g., ferric sulphate and ferric chloride).

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## Solubilizing and suspending agents

Suitable suspending agents can include, for example, ethoxylated isostearyl alcohols, polyoxyethylene sorbitol and sorbitan esters, microcrystalline cellulose, aluminum metahydroxide, bentonite, agar-agar and tragacanth, or mixtures of these substances, and the like.

The disclosed antimicrobial compositions can also comprise solubilizing agents and emulsifiers, as for example, ethyl alcohol, isopropyl alcohol, ethyl carbonate; ethyl acetate, benzyl alcohol, benzyl alcohol, benzyl benzoate, propyleneglycol, 1,3-butyleneglycol, dimethylformamide, oils, in particular, cottonseed oil, groundnut oil, corn germ oil, olive oil, castor oil and sesame oil, glycerol, tetrahydrofurfuryl alcohol, polyethyleneglycols and fatty acid esters of sorbitan or mixtures of these substances, and the like.

## Additional Quaternary Ammonium Salts

In one aspect, the disclosed antimicrobial compositions can comprise one or more additional quaternary ammonium salts. Other additional quaternary ammonium salts that can be used in the disclosed antimicrobial compositions include, but are not limited to, other aliphatic heteroaryl salts (e.g., alkyl pyridinium halides, alkyl quinolinium halides, alkyl indolinium halides, and the like), aliphatic heterocyclic salts (e.g., aliphatic heterocycloalkyl salts like alkyl piperidinium salts or aliphatic heterocycloalkenyl salts), aliphatic benzylalkyl ammoniums salts, dialiphatic dialkyl ammoniums salts, and tetraalkyl ammonium salts, and chloramine-T.

#### **Amounts**

The additional components disclosed herein can be present in the disclosed antimicrobial compositions in any amount as is described above for the aliphatic benzylalkyl ammonium salts, dialiphatic dialkyl ammonium salts, tetraalkyl ammoniums salts, and/or trichlormelamine. For example, one or more additional components can be present in an amount of from about 0.001 to about 0.1 weight %, from about 0.005 to about 0.075 weight %, from about 0.0075 about 0.05 weight %, from about 0.01 to about 0.02 weight %, about 0.005 to about 0.1 weight %, about 0.005 to about 0.02 weight %, or about 0.01 weight %, based on the total weight of the antimicrobial composition. In another example, the disclosed antimicrobial compositions can contain from about 0.001 to about 0.1 parts by weight, from about 0.005 to about 0.075 parts by weight, from about 0.0075 about 0.05 parts by weight, from

about 0.01 to about 0.02 parts by weight, about 0.005 to about 0.1 parts by weight, about 0.005 to about 0.02 parts by weight, about 0.005 to about 0.01 parts by weight, or about 0.01 parts by weight, based of one or more additional components.

# **Exemplary Compositions**

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In one example, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, and a tetraalkyl ammonium salt. For example, disclosed herein are antimicrobial compositions that consist essentially of an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, and a tetraalkyl ammonium salt. "Consisting essentially of" is used herein to exclude additional components or the omission of components that would change the basic and novel characteristics of the composition; this is also meant to exclude the omission of aliphatic heteroaryl salts, trichloromelamine, aliphatic benzylalkyl ammonium salts, dialiphatic dialkyl ammoniums salts, and/or tetraalkyl ammonium salts from the composition but not the addition or omission of other carriers, adjuvants, solubilizing and suspending agents, and additional components as described herein. The composition can also comprise water.

In a further example, disclosed herein in an antimicrobial composition comprising an aliphatic heteroaryl salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt. For example, disclosed herein are antimicrobial compositions that consist essentially of an aliphatic heteroaryl salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt.

In a still further example, disclosed herein in an antimicrobial composition comprising an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, and a dialiphatic dialkyl ammonium salt. For example, disclosed herein are antimicrobial compositions that consist essentially of an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, and a dialiphatic dialkyl ammonium salt.

In the disclosed compositions, the aliphatic heteroaryl salt can be any aliphatic heteroaryl salt disclosed herein, for example, an alkylpyridinium halide. Such an alkylpyridinium halide can comprise, for example, cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof. The aliphatic benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein, for example, an alkyl dimethyl benzyl ammonium bromide, or a mixture thereof. The dialiphatic dialkyl ammonium salt can be any dialiphatic dialkyl

ammonium salt disclosed herein, for example, di-dodecyl dimethyl ammonium chloride or bromide, di-tetradecyl dimethyl ammonium chloride or bromide, dihexadecyl dimethyl ammonium chloride or bromide, or mixtures thereof. The tetraalkyl ammonium salt can be any tetraalkyl ammonium salt disclosed herein, for example, cetyl trimethyl ammonium chloride or bromide, lauryl trimethyl ammonium chloride or bromide, myristyl trimethyl ammonium chloride or bromide, or mixtures thereof.

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In one example, disclosed herein are antimicrobial compositions that comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, and at least two ammonium salts comprising an aliphatic heteroaryl salt, a dialiphatic dialkyl ammonium salt, or a tetraalkyl ammonium salt. In another example, disclosed herein are antimicrobial compositions that comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, a tetraalkyl ammonium salt, and an ammonium salt comprising an aliphatic heteroaryl salt or a dialiphatic dialkyl ammonium salt. In yet another example, disclosed herein are antimicrobial compositions that comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, a dialiphatic dialkyl ammonium salt, and an ammonium salt comprising an aliphatic heteroaryl salt or a tetraalkyl ammonium salt. In still another example, disclosed herein are antimicrobial compositions that comprise an aliphatic benzylalkyl ammonium salt, trichloromelamine, an aliphatic heteroaryl salt, and an ammonium salt comprising a tetraalkyl ammonium salt or a dialiphatic dialkyl ammonium salt.

In these compositions, when the aliphatic heteroaryl salt is present it can be as disclosed above; for example, it can comprise an alkylpyridinium halide as disclosed herein (e.g., cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof). The aliphatic benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein (e.g., alkyl dimethyl benzyl ammonium halide, alkyl dimethyl benzyl ammonium halide, or a mixture thereof). When the dialiphatic dialkyl ammonium salt is present, it can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof). When the tetraalkyl ammonium salt is present, it can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, or a mixture thereof).

In these compositions, when the two ammonium salts are an aliphatic heteroaryl

salt and a tetraalkyl ammonium salt, the aliphatic heteroaryl salt can be any aliphatic heteroaryl salt disclosed herein (e.g., cetyl pyridinium halide) and the tetraalkyl ammonium salt can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof). When the two ammonium salts are an aliphatic heteroaryl salt and a dialiphatic dialkyl ammonium salt, the aliphatic heteroaryl salt can be any aliphatic heteroaryl salt disclosed herein (e.g., cetyl pyridinium halide) and the dialiphatic dialkyl ammonium salt can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof). When the two ammonium salts are a dialiphatic dialkyl ammonium salt and a tetraalkyl ammonium salt, the dialiphatic dialkyl ammonium salt can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof) and the tetraalkyl ammonium salt can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof).

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The composition can contain the aliphatic heteroaryl salt in any of the amounts disclosed above. For example, the aliphatic benzylalkyl ammonium salt can be present in an amount of from about 3.5 to about 8 parts by weight. The composition can contain trichloromelamine, the aliphatic heteroaryl salt, the dialiphatic dialkyl ammonium salt, or the tetraalkyl ammonium salt in any of the amounts disclosed above. For example, trichloromelamine, aliphatic heteroaryl salt, dialiphatic dialkyl ammonium salt, or tetraalkyl ammonium salt can be present in an amount of from about 0.001 to about 0.1 parts by weight. It is also contemplated that these compositions can further comprise water.

A suitable antimicrobial composition can comprise an aliphatic benzylalkyl ammonium salt in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight). In another example, an antimicrobial composition can comprise trichloromelamine in an amount of from about 0.001 to about 1.0 weight % (or from about 0.001 to about 1.0 parts by weight). In another example, an antimicrobial composition can comprise an aliphatic heteroaryl salt in an amount of from about 0.001 to

about 1.0 weight % (or from about 0.001 to about 1.0 parts by weight). In another example, an antimicrobial composition can comprise a dialiphatic dialkyl ammonium salt in an amount of from about 0.001 to about 1.0 weight % (or from about 0.001 to about 1.0 parts by weight). In another example, an antimicrobial composition can comprise a tetraalkyl ammonium salt in an amount of from about 0.001 to about 1.0 weight % (or from about 0.001 to about 1.0 parts by weight). And in another example, an antimicrobial composition can optionally comprise an additional component in an amount of from about 0.001 to about 1.0 weight % (or from about 0.001 to about 1.0 parts by weight). In another example, the aliphatic heteroaryl salt can be present in an amount of from about 3.5 to about 8 parts by weight, the trichloromelamine can be present in an amount of from about 0.001 to about 1.0 parts by weight, and the other two ammonium salts can each be present in an amount of from about 0.001 to about 1.0 parts by weight.

In a specific example, the disclosed composition can comprise an aliphatic benzylalkyl ammonium salt such as alkyl dimethyl benzyl ammonium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.5 parts by weight, and two ammonium salts such as an aliphatic heteroaryl salt present in an amount of from about 0.005 to about 0.5 parts by weight and a tetraalkyl ammonium salt present in an amount of from about 0.005 to about 0.5 parts by weight. In a specific example, the amount of alkyl dimethyl benzyl ammonium halide can be about 7.5 parts by weight, the amount of trichloromelamine can be about 0.1 parts by weight, the amount of cetylpyridinium chloride can be about 0.08 parts by weight, and the amount of cetyl trimethyl ammonium chloride can be about 0.05 parts by weight.

In another specific example, disclosed are compositions comprising an aliphatic benzylalkyl ammonium salt such as alkyl dimethyl benzyl ammonium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.5 parts by weight, and two ammonium salts such as an aliphatic heteroaryl salt present in an amount of from about 0.005 to about 0.5 parts by weight and a dialiphatic dialkyl ammonium salt present in an amount of from about 0.005 to about 0.5 parts by weight. In yet another specific example, the disclosed compositions can comprise an aliphatic benzylalkyl ammonium salt such as alkyl dimethyl benzyl ammonium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.5 parts by weight, and two ammonium salts such as a dialiphatic dialkyl ammonium salt

present in an amount of from about 0.005 to about 0.5 parts by weight and a tetraalkyl ammonium salt present in an amount of from about 0.005 to about 0.5 parts by weight.

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In another aspect, disclosed herein is an antimicrobial composition comprising alkyl pyridinium halide, trichloromelamine, alkyl benzylalkyl ammonium halide, and tetraalkyl ammonium halide. For example, a suitable antimicrobial composition can comprise alkyl pyridinium halide in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight), trichloromelamine in an amount of from about 0.005 to about 0.5 weight % (or from about 0.005 to about 0.5 parts by weight), alkyl benzylalkyl ammonium halide in an amount of from about 0.005 to about 0.5 weight % (or from about 0.005 to about 0.5 parts by weight), and a tetraalkyl ammonium halide in an amount of from about 0.005 to about 0.5 parts by weight), and a balance of water.

In still another aspect, disclosed herein are antimicrobial compositions that comprise cetylpyridinium halide, trichloromelamine, alkyl benzylalkyl ammonium halide, and tetraalkyl ammonium halide. For example, a suitable antimicrobial composition can comprise cetylpyridinium chloride in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight) or from about 6 to about 8 weight % (or from about 6 to about 8 parts by weight), trichloromelamine in an amount of from about 0.005 to about 0.5 weight % (or from about 0.005 to about 0.5 parts by weight), alkyl dimethyl benzyl ammonium chloride and/or alky methylethyl benzyl ammonium chloride in an amount of from about 0.005 to about 0.5 weight % (or from about 0.005 to about 0.5 parts by weight), and cetyl trimethyl ammonium chloride in an amount of from about 0.005 to about 0.5 parts by weight), and a balance of water. Another suitable example involves the use of the bromide salts of the previous composition.

In and other example, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt and trichloromelamine. For examples, disclosed herein are antimicrobial compositions that consist essentially of an aliphatic heteroaryl salt and trichloromelamine. In one aspect, the antimicrobial composition does not contain aliphatic benzylalkyl ammonium salts, dialiphatic dialkyl ammoniums salt, and tetraalkyl ammonium salts. The composition can also comprise water.

In the disclosed compositions, the aliphatic heteroaryl salt can be any aliphatic heteroaryl salt disclosed herein, for example, an alkylpyridinium halide. Such as alkylpyridinium halide can comprise cetylpyridinium chloride, cetylpyridinium bromide,

or a mixture thereof. The aliphatic benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein, for example, an alkyl dimethyl benzyl ammonium chloride, alkyl dimethyl benzyl ammonium bromide, or a mixture thereof.

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The composition can contain the aliphatic heteroaryl salt in any of the amounts disclosed above. For example, the aliphatic heteroaryl salt can be present in an amount of from about 3.5 to about 8 parts by weight. The composition can contain trichloromelamine in any of the amounts disclosed above. For example, trichloromelamine can be present in an amount of from about 0.005 to about 0.02 parts by weight.

In another aspect, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt, trichloromelamine; and an ammonium salt selected from the group consisting of an aliphatic benzylalkyl ammonium salt, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt. In these compositions, when the ammonium salt is the aliphatic benzyl ammonium salt, the composition does not contain the dialiphatic dialkyl ammonium salt or the tetraalkyl ammonium salt. Alternatively, when the ammonium salt is the dialiphatic dialkyl ammonium salt, the composition does not contain the aliphatic benzyl ammonium salt or the tetraalkyl ammonium salt. Also, when the ammonium salt is the tetraalkyl ammonium salt, the composition does not contain the aliphatic benzyl ammonium salt or the dialiphatic dialkyl ammonium salt. Also disclosed are compositions that consist essential of an aliphatic heteroaryl salt, trichloromelamine, and an ammonium salt selected from the group consisting of an aliphatic benzylalkyl ammonium salt, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt. It is also contemplated that these compositions can further comprise water.

In one example, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt. In another aspect, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, and water. For example, a suitable antimicrobial composition can comprise an aliphatic heteroaryl salt in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight). In another example, an antimicrobial composition can comprise an aliphatic benzylalkyl ammonium salt in an amount of from about 0.005 to about 0.1 weight % (or from about 0.005 to about 0.1 parts by weight). In another example, an antimicrobial composition can comprise trichloromelamine in an amount of from about

0.005 to about 0.02 weight % (or from about 0.005 to about 0.02 parts by weight). And in another example, an antimicrobial composition can optionally comprise an additional component in an amount of from about 0.005 to about 0.02 weight % (or from about 0.005 to about 0.02 parts by weight).

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In these compositions, the aliphatic heteroaryl salt can be as disclosed above; for example, it can comprise an alkylpyridinium halide as disclosed herein (e.g., cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof). When the ammonium salt is the aliphatic benzylalkyl ammonium salt, it can be any aliphatic benzylalkyl ammonium salt disclosed herein (e.g., alkyl dimethyl benzyl ammonium halide, alkyl dimethyl benzyl ammonium halide, or a mixture thereof). When the ammonium salt is the dialiphatic dialkyl ammonium salt, it can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof). When the ammonium salt is the tetraalkyl ammonium salt, it can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof).

The amounts of these components can be as described before. For example, the aliphatic heteroaryl salt can be present in an amount of from about 3.5 to about 8 parts by weight, the trichloromelamine can be present in an amount of from about 0.005 to about 0.02 parts by weight, and the ammonium salt can be present in an amount of from about 0.005 to about 0.1 parts by weight.

A specific example of these compositions includes the composition comprising an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, an aliphatic benzylalkyl ammonium salt such as alkyl dimethyl benzyl ammonium chloride present in an amount of from about 0.005 to about 0.02 parts by weight. In one example, the composition does not contain a dialiphatic dialkyl ammonium salt or a tetraalkyl ammonium salt.

Another example includes the composition comprising an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and an aliphatic benzylalkyl ammonium salt such as alkyl methylethyl benzyl ammonium chloride present in an amount of from about 0.005 to about 0.1 parts

by weight. In one example, the composition does not contain a dialiphatic dialkyl ammonium salt or a tetraalkyl ammonium salt.

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Yet another example includes the composition comprising an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and a dialiphatic dialkyl ammonium salt such as didodecyl dimethyl ammonium chloride present in an amount of from about 0.005 to about 0.1 parts by weight. In one example, the composition does not contain an aliphatic benzylalkyl ammonium salt or a tetraalkyl ammonium salt.

A further example includes the composition comprising an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and a tetraalkyl ammonium salt such as cetyl dimethyl ammonium chloride present in an amount of from about 0.005 to about 0.1 parts by weight. In one example, the composition does not contain a dialiphatic dialkyl ammonium salt or an aliphatic benzylalkyl ammonium salt.

In yet another aspect, disclosed herein are antimicrobial compositions that comprise an aliphatic heteroaryl salt, trichloromelamine, and two ammonium salts selected from the group consisting of an aliphatic benzylalkyl ammonium salt and a dialiphatic dialkyl ammonium salt, a aliphatic benzylalkyl ammonium salt and a tetraalkyl ammonium salt, or a dialiphatic dialkyl ammonium salt and a tetraalkyl ammonium salt. It is contemplated that these compositions can further comprise water. As with the other compositions, the aliphatic heteroaryl salt component can be any aliphatic heteroaryl salt described above (e.g., an alkylpyridinium halide such as cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof).

In these compositions, when the two ammonium salts are an aliphatic benzylalkyl ammonium salt and a tetraalkyl ammonium salt, the aliphatic benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein (e.g., alkyl dimethyl benzyl ammonium halide, alkyl dimethyl benzyl ammonium halide, or a mixture thereof) and the tetraalkyl ammonium salt can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof). When the two ammonium salts are an aliphatic benzylalkyl ammonium salt and a dialiphatic dialkyl ammonium salt, the aliphatic

benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein (e.g., alkyl dimethyl benzyl ammonium halide, alkyl dimethyl benzyl ammonium halide, or a mixture thereof) and the dialiphatic dialkyl ammonium salt can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof). When the two ammonium salts are a dialiphatic dialkyl ammonium salt and a tetraalkyl ammonium salt, the dialiphatic dialkyl ammonium salt can be any dialiphatic dialkyl ammonium salt disclosed herein (e.g., didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof) and the tetraalkyl ammonium salt can be any tetraalkyl ammonium salt disclosed herein (e.g., cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof).

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In these compositions, the aliphatic heteroaryl salt can be present in any amount as disclosed above; for example, it can be present in an amount of from about 3.5 to about 8 parts by weight. The trichloromelamine can also be present in any amount disclosed herein, such as from about 0.005 to about 0.02 parts by weight. Further, the two ammonium salts can each be present in any amount as disclosed herein (e.g., from about 0.005 to about 0.1 parts by weight).

In a specific example, the disclosed composition can comprise an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and two ammonium salts such as an aliphatic benzylalkyl ammonium salt present in an amount of from about 0.005 to about 0.1 parts by weight and a tetralkyl ammonium salt present in an amount of from about 0.005 to about 0.02 parts by weight. In a specific example, the amount of cetylpyridinium chloride can be about 7.5 parts by weight, the amount of trichloromelamine can be about 0.01 parts by weight, the amount of alkyl dimethyl benzyl ammonium chloride can be about 0.01 parts by weight, and the amount of cetyl trimethyl ammonium chloride can be about 0.01 parts by weight.

In another specific example, disclosed are compositions comprising an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and two ammonium salts such as an aliphatic benzylalkyl

ammonium salt present in an amount of from about 0.005 to about 0.1 parts by weight and a dialiphatic dialkyl ammonium salt present in an amount of from about 0.005 to about 0.02 parts by weight. In yet another specific example, the disclosed compositions can comprise an aliphatic heteroaryl salt such as cetylpyridinium chloride present in an amount of from about 3.5 to about 8 parts by weight, trichloromelamine present in an amount of from about 0.005 to about 0.02 parts by weight, and two ammonium salts such as a dialiphatic dialkyl ammonium salt present in an amount of from about 0.005 to about 0.1 parts by weight and a tetralkyl ammonium salt present in an amount of from about 0.005 to about 0.005 to about 0.002 parts by weight.

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Suitable examples of aliphatic benzylalkyl ammonium salts are disclosed herein and include, for example, alkyl dimethyl benzyl ammonium chloride. Suitable examples of tetraalkyl ammonium salts are disclosed herein and include, for example, cetyl trimethyl ammonium chloride. Suitable examples of dialiphatic dialkyl ammonium salts are disclosed herein and include, for example, didodecyl dimethyl ammonium chloride.

In another aspect, disclosed herein are antimicrobial compositions that comprise alkyl pyridinium halide, alkyl benzylalkyl ammonium halide, and trichloromelamine. For example, a suitable antimicrobial composition can comprise alkyl pyridinium halide in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight), alkyl benzylalkyl ammonium halide in an amount of from about 0.005 to about 0.1 weight % (or from about 0.005 to about 0.1 parts by weight), trichloromelamine in an amount of from about 0.005 to about 0.02 weight % (or from about 0.005 to about 0.02 parts by weight), and optionally a tetralkyl ammonium halide in an amount of from about 0.005 to about 0.02 parts by weight), and a balance of water.

In still another aspect, disclosed herein are antimicrobial compositions that comprise cetylpyridinium halide, alkyl benzylalkyl ammonium halide, and trichloromelamine. For example, a suitable antimicrobial composition can comprise cetylpyridinium chloride in an amount of from about 3.5 to about 8 weight % (or from about 3.5 to about 8 parts by weight) or from about 6 to about 8 weight % (or from about 6 to about 8 parts by weight), alkyl dimethyl benzyl ammonium chloride and/or alky methylethyl benzyl ammonium chloride in an amount of from about 0.005 to about 0.1 weight % (or from about 0.005 to about 0.1 parts by weight), trichloromelamine in an amount of from about 0.005 to about 0.02 weight % (or from about 0.005 to about 0.02 parts by weight), and cetyl trimethyl ammonium chloride in an amount of from about

0.005 to about 0.02 weight % (or from about 0.005 to about 0.02 parts by weight), and a balance of water. Another suitable example involves the use of the bromide salts of the previous composition.

In yet another aspect, a suitable antimicrobial composition can comprise 7.5 weight % (from about 7.5 parts by weight) of alkyl pyridinium halide (e.g., cetylpyridinium chloride (and/or bromide), 0.01 weight % (or 0.01 part by weight) of aliphatic benzylalkyl ammonium halide (e.g., cetyl dimethyl benzyl ammonium chloride and/or bromide), 0.01 weight % (or 0.01 part by weight) of trichloromelamine, and optionally 0.01 weight % (or 0.01 part by weight) of an additional component (e.g., cetyl trimethyl ammonium chloride and/or bromide) and a balance of water.

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In yet another aspect, the disclosed antimicrobial compositions can comprise about 7.5 parts of aliphatic heteroaryl salt (e.g., cetylpyridinium chloride), 0.1 part of aliphatic benzyalkyl ammonium salt (e.g., cetyl dimethyl benzyl ammonium chloride), 0.1 part of trichloromelamine, and optionally 0.1 part of cetyl trimethyl ammonium chloride, and balance water (e.g., 92.2 parts).

In still another aspect, disclosed are aqueous compositions that comprise effective amounts of a combination of at least two quaternary ammonium salts, an ammonium halide, trichlormelamine, and water. The combination of at least two quaternary ammonium salts is selected from the group consisting of cetyl pyridinium chloride, *N*-alkyl dimethyl benzyl ammonium chloride, and alkyl dimethyl ethyl benzyl ammonium chloride. The combination of at least two quaternary ammonium salts is present in an amount of about 6.02 to 8.02 weight percent.

In a further aspect, disclosed herein are compositions that contain an aliphatic heteroaryl salt and trichloromelamine. The amount of these components can be as described above.

Still further, a suitable antimicrobial composition can contain an aliphatic heteroaryl salt, trichloromelamine, and a tetraalkyl ammonium salt. The amount of these components in the composition can be as described above.

In and other example, disclosed herein are antimicrobial compositions that comprise an aliphatic benzylalkyl ammonium salt and trichloromelamine. For examples, disclosed herein are antimicrobial compositions that consist essentially of an aliphatic benzylalkyl ammonium salt and trichloromelamine. In one aspect, the antimicrobial composition does not contain aliphatic heteroaryl salts, dialiphatic dialkyl ammonium salt, and tetraalkyl ammonium salts. The composition can also comprise water.

In the disclosed compositions, the aliphatic benzylalkyl ammonium salt can be any aliphatic benzylalkyl ammonium salt disclosed herein, for example, an alkyl benzalkonium halide such as alkyl dimethyl benzyl ammonium chloride, which is a mixture of C12-C18 alkyl dimethyl benzyl ammonium chloride, and alkyl methylethyl benzyl ammonium bromide, including mixtures thereof. The composition can contain the aliphatic benzylalkyl ammonium salt in any of the amounts disclosed above. For example, the aliphatic benzylalkyl ammonium salt can be present in an amount of from about 3.5 to about 8 parts by weight. The composition can contain trichloromelamine in any of the amounts disclosed above. For example, trichloromelamine can be present in an amount of from about 0.005 to about 0.02 parts by weight.

#### **Forms**

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Depending on the intended mode of use, as is discussed below, the antimicrobial compositions disclosed herein can be in the form of solid, semi-solid, liquid, or gel forms, such as, for example, tablets, pills, capsules, powders, liquids, suspensions, dispersions, or emulsions. Also, the compositions disclosed herein can be in a form suitable for dilution. That is, the compositions can be in the form of an aqueous or non-aqueous stock solution, concentrate, concentrated solution, dispersion, emulsion, or suspension that can be diluted to a desired concentration with a suitable solvent (e.g., water). Similarly, the compositions can be in the form of a powder, paste, cream, or solid that can be reconstituted or mixed with a solvent and diluted to a desired concentration to form a solution or dispersion, emulsion, emulsifiable concentrated, slurries, or suspension. In one example, the disclosed antimicrobial compositions can be in the form of a solution, such as an aqueous solution.

Contacting aqueous environments with the disclosed antimicrobial compositions will depend on the system and form of the compositions. For example, the compositions can be added to the aqueous environment by pouring in a concentrate of the composition. Diluted solutions of the composition, as are described herein, can also be poured into the aqueous environment. Further, a tablet or cake of the disclosed compositions can be added to the aqueous environment and dissolved. Still further, automatic dispensing apparatus can be used to supply the disclosed compositions at regular intervals or continuously. In general, any method for incorporating a liquid or dissolved solid into an aqueous medium can be used herein. Mixing the aqueous environment before, during, and/or after the composition is incorporated will often be desired. Also, incorporating the disclosed compositions can be done at any stage of a process; the particular stage

depending upon the desires of the practitioner. For a pool, as example, the disclosed compositions can be added to the pool water at the pre-filter and/or post-filter stage.

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It has been found that the disclosed antimicrobial compositions are equally effective even when concentrated or when diluted with water up to a certain point. For example, it has been found that the disclosed antimicrobial compositions can be diluted with water in the range of about 1 to about 400 parts water to one part antimicrobial composition and still perform effectively. In some specific examples, the antimicrobial compositions disclosed herein can be diluted with water in a ratio of about 1:1, 2:1, 3:1, 4:1, 5:1, 6:1, 7:1, 8:1, 9:1, 10:1, 11:1, 12:1, 13:1, 14:1, 15:1, 16:1, 17:1, 18:1, 19:1, 20:1, 21:1, 22:1, 23:1, 24:1, 25:1, 26:1, 27:1, 28:1, 29:1, 30:1, 31:1, 32:1, 33:1, 34:1, 35:1, 10 36:1, 37:1, 38:1, 39:1, 40:1, 41:1, 42:1, 43:1, 44:1, 45:1, 46:1, 47:1, 48:1, 49:1, 50:1, 51:1, 52:1, 53:1, 54:1, 55:1, 56:1, 57:1, 58:1, 59:1, 60:1, 61:1, 62:1, 63:1, 64:1, 65:1, 66:1, 67:1, 68:1, 69:1, 70:1, 71:1, 72:1, 73:1, 74:1, 75:1, 76:1, 77:1, 78:1, 79:1, 80:1, 81:1, 82:1, 83:1, 84:1, 85:1, 86:1, 87:1, 88:1, 89:1, 90:1, 91:1, 92:1, 93:1, 94:1, 95:1, 96:1, 97:1, 98:1, 99:1, 100:1, 101:1, 102:1, 103:1, 104:1, 105:1, 106:1, 107:1, 108:1, 15 109:1, 110:1, 111:1, 112:1, 113:1, 114:1, 115:1, 116:1, 117:1, 118:1, 119:1, 120:1, 121:1, 122:1, 123:1, 124:1, 125:1, 126:1, 127:1, 128:1, 129:1, 130:1, 131:1, 132:1, 133:1, 134:1, 135:1, 136:1, 137:1, 138:1, 139:1, 140:1, 141:1, 142:1, 143:1, 144:1, 145:1, 146:1, 147:1, 148:1, 149:1, 150:1, 151:1, 152:1, 153:1, 154:1, 155:1, 156:1, 157:1, 158:1, 159:1, 160:1, 161:1, 162:1, 163:1, 164:1, 165:1, 166:1, 167:1, 168:1, 169:1, 170:1, 171:1, 172:1, 173:1, 20 174:1, 175:1, 176:1, 177:1, 178:1, 179:1, 180:1, 181:1, 182:1, 183:1, 184:1, 185:1, 186:1, 187:1, 188:1, 189:1, 190:1, 191:1, 192:1, 193:1, 194:1, 195:1, 196:1, 197:1, 198:1, 199:1, 200:1, 201:1, 202:1, 203:1, 204:1, 205:1, 206:1, 207:1, 208:1, 209:1, 210:1, 211:1, 212:1, 213:1, 214:1, 215:1, 216:1, 217:1, 218:1, 219:1, 220:1, 221:1, 222:1, 223:1, 224:1, 225:1, 226:1, 227:1, 228:1, 229:1, 230:1, 231:1, 232:1, 233:1, 234:1, 235:1, 236:1, 237:1, 238:1, 25 239:1, 240:1, 241:1, 242:1, 243:1, 244:1, 245:1, 246:1, 247:1, 248:1, 249:1, 250:1, 251:1, 252:1, 253:1, 254:1, 255:1, 256:1, 257:1, 258:1, 259:1, 260:1, 261:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 262:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 264:1, 263:1, 263:1, 264:1, 264:1,265:1, 266:1, 267:1, 268:1, 269:1, 270:1, 271:1, 272:1, 273:1, 274:1, 275:1, 276:1, 277:1, 278:1, 279:1, 280:1, 281:1, 282:1, 283:1, 284:1, 285:1, 286:1, 287:1, 288:1, 289:1, 290:1, 280:1,291:1, 292:1, 293:1, 294:1, 295:1, 296:1, 297:1, 298:1, 299:1, 300:1, 301:1, 302:1, 303:1, 30 304:1, 305:1, 306:1, 307:1, 308:1, 309:1, 310:1, 311:1, 312:1, 313:1, 314:1, 315:1, 316:1, 317:1, 318:1, 319:1, 320:1, 321:1, 322:1, 323:1, 324:1, 325:1, 326:1, 327:1, 328:1, 329:1, 330:1, 331:1, 332:1, 333:1, 334:1, 335:1, 336:1, 337:1, 338:1, 339:1, 340:1, 341:1, 342:1, 343:1, 344:1, 345:1, 346:1, 347:1, 348:1, 349:1, 350:1, 351:1, 352:1, 353:1, 354:1, 355:1,

356:1, 357:1, 358:1, 359:1, 360:1, 361:1, 362:1, 363:1, 364:1, 365:1, 366:1, 367:1, 368:1, 369:1, 370:1, 371:1, 372:1, 373:1, 374:1, 375:1, 376:1, 377:1, 378:1, 379:1, 380:1, 381:1, 382:1, 383:1, 384:1, 385:1, 386:1, 387:1, 388:1, 389:1, 390:1, 391:1, 392:1, 393:1, 394:1, 395:1, 396:1, 397:1, 398:1, 399:1, or 400:1 parts water to parts antimicrobial composition; these ratios can also be an upper and lower endpoint of a range of ratios when appropriate.

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It has also been discovered that much higher dilutions can be used for certain applications. For example, dilutions of from about 1 to 500 up to about 1 to 1000 can be used for drinking water (human or animal).

The disclosed antimicrobial compositions are still be effective when present in a solution at from about 20 to about 500 parts per million (ppm), or from about 20 to about 200 ppm, based on the aliphatic heteroaryl salt component. For example, the disclosed antimicrobial compositions can be in a solution at about 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140, 150, 160, 170, 180, 190, 200, 210, 220, 230, 240, 250, 260, 270, 280, 290, 300, 310, 320, 330, 340, 350, 360, 370, 380, 390, 400, 410, 420, 430, 440, 450, 460, 470, 480, 490, 500 ppm or more, based on the aliphatic heteroaryl salt component, where any of the stated values can form an upper or lower endpoint when appropriate. In some particular aspects, the disclosed compositions can be effective at concentrations of at or below about 100 ppm (e.g., at or below 50 ppm).

It is expected that a dosage of the disclosed compositions as a liquid can be about 7.5 ppm to initially clean up a pool then about 1.5 ppm once per week to maintain control. For cooling towers, the disclosed compositions can be applied in liquid form to bulk recirculation water at dosages from about 0.5 to about 50 ppm as needed to achieve desired microbiological control. For paper machines, the disclosed compositions can be applied in liquid form to white water system or fresh water make-up at from about 1 to about 1000 ppm to achieve desired control. The disclosed compositions can also be added to starch systems and to mineral additive systems as a preservative at dosages of from about 20 to about 1000 ppm. In aquaculture applications, as described herein, the disclosed compositions can be used to control bacteria, fungi, and algae at dosage ranges from about 0.015 to about 0.1 ppm for bulk water and from about 1 to about 5 ppm for dips and baths to remove and/or control parasites. For oil field waters, the disclosed compositions can be used in a dosage range of from about 1 to about 1500 ppm. It is understood, of course, that the exact dosage swill depend on the conditions of the aqueous

environment (e.g., volume, degree of contamination, location), but optimization of the dose can be performed by the skilled artisan.

# Methods of Making

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Certain materials, compounds, compositions, and components disclosed herein can be obtained commercially or can be readily synthesized using techniques generally known to those of skill in the art. For example, the starting materials and reagents used in preparing the disclosed compounds and compositions are either available from commercial suppliers such as Aldrich Chemical Co., (Milwaukee, Wis.), Acros Organics (Morris Plains, N.J.), Fisher Scientific (Pittsburgh, Pa.), or Sigma (St. Louis, Mo.) or are prepared by methods known to those skilled in the art following procedures set forth in references such as Fieser and Fieser's Reagents for Organic Synthesis, Volumes 1-17 (John Wiley and Sons, 1991); Rodd's Chemistry of Carbon Compounds, Volumes 1-40 (John Wiley and Sons, 1991); March's Advanced Organic Reactions, Volumes 1-40 (John Wiley and Sons, 1991); March's Advanced Organic Chemistry, (John Wiley and Sons, 4th Edition); and Larock's Comprehensive Organic Transformations (VCH Publishers Inc., 1989). Alternatively, the components used in the antimicrobial compositions disclosed herein can be purchased from commercial suppliers.

The disclosed antimicrobial compositions can be prepared by admixing, in any order, an aliphatic heteroaryl salt, trichloromelamine, and one or more of an aliphatic benzylalkyl ammonium salt, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt, and any optional additional components. Also, disclosed is an antimicrobial composition prepared by such a method. The resulting composition can also be diluted as described herein.

#### **EXAMPLES**

The following examples are set forth below to illustrate the methods and results according to the disclosed subject matter. These examples are not intended to be inclusive of all aspects of the subject matter disclosed herein, but rather to illustrate representative methods and results. These examples are not intended to exclude equivalents and variations of the present invention which are apparent to one skilled in the art.

Efforts have been made to ensure accuracy with respect to numbers (e.g., amounts, temperature, etc.) but some errors and deviations should be accounted for.

Unless indicated otherwise, parts are parts by weight, temperature is in °C or is at ambient temperature, and pressure is at or near atmospheric. There are numerous variations and

combinations of reaction conditions, e.g., component concentrations, desired solvents, solvent mixtures, temperatures, pressures and other reaction ranges and conditions that can be used to optimize the product purity and yield obtained from the described process. Only reasonable and routine experimentation will be required to optimize such process conditions.

## Example 1

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The effects of an antimicrobial composition as disclosed herein were studied on pathogenic, indicator, and spoilage populations of bacteria associated with broiler chicken carcasses. Scalder water was collected from the overflow end (the entrance end) of a commercial poultry scalder. The water was autoclaved to eliminate all populations of bacteria and bacterial spores to avoid interference during the study. The autoclaved scalder water was evaluated chemically and compared to raw scalder water to ensure that the organic material in raw and autoclaved scalder water was similar.

A test solution (interchangeably referred to in the examples as the antimicrobial composition) was prepared. The test solution contained cetylpyridinium chloride (7.5 parts by weight), alkyl dimethyl benzyl ammonium chloride (0.1 part by weight), trichloromelamine (0.1 part by weight), cetyl trimethyl ammonium chloride (0.1 part by weight), and water (92.2 parts by weight). Next, a control solution was prepared by admixing cetylpyridinium chloride (7.5 parts by weight) and water (92.5 parts by weight). The same solutions were used in all of the examples.

Sets of test tubes were prepared by adding 9 mL of autoclaved (sterilized) scalder water to sterile polystyrene test tubes. One set was prepared as controls by adding 9 mL of autoclaved scalder water to tubes. Another set was prepared by adding 9 mL of autoclaved scalder water and 1 mL of the test solution as identified above. The pathogens were Salmonella typhimurium ("ST"), Listeria monocytogenes ("LM"), and Staphylococcus aureau ("SA"). The indicator was Escherichia coli ("EC") and the spoilage bacteria were Pseudomonas fluorscens ("PF") and Shewanella putrefaciens ("SP"). These microorganisms were grown overnight in Brian Heart infusion broth at 25 °C for 24 hours. Each bacterium was exposed to each autoclaved scalder watersanitizer combination for 2 minutes to mimic scalding. After exposure period, 1 mL of the suspension was placed into 9 mL of Brian Heart infusion broth and vortexed. One mL of this mixture was placed into the Bactometer module and bacterial growth was measured. The results are provided at Figures 1-4.

It can be seen from Figure 1 that the antimicrobial composition disclosed herein was effective for reducing populations of Salmonella, Listeria, Staphylococcus, and Shewanella when used in combination with scalder water applications. In the meantime, a substantial reduction is seen for Escherichia coli and Pseudomonas fluorescens. In comparison, the control solution eliminated much less of any of the above microorganisms.

Figure 2 is a graph that comparatively shows the reduction of bacterial colonies when exposed to a solution as disclosed herein and a solution of only cetylpyridinium chloride. The colony forming units for Salmonella typhimurium, Listeria monocytogenes, Staphylococcus aureau, and Escherichia coli were tested. Although not depicted with Logic CFU in Figure 2, Pseudomonas was also reduced to below 10 CFU/mL.

Figure 3 is a graph showing the effect of the test solution as compared with the control solution. It can be seen from Figure 3 that over a period of 24 hours, Salmonella typhimurium, Listeria monocytogenes, Staphylococcus aureus, and Shewanella putrefaciens were completely eliminated while E. coli and Pseudomonas fluorscens were substantially reduced as compared with samples treated with the control solution.

Figure 4 is a graph that comparatively shows the reduction of bacterial colonies when exposed to the test solution and the control solution. Figure 4 is similar to Figure 2 and shows that the colony forming units for all microorganisms where nearly eliminated upon treatment with the antimicrobial test solution. Thus, the antimicrobial solution was effective in eliminating all pathogenic, indicator, and spoilage bacteria tested in combination with scalder water applications. This data also indicates effectiveness of the test solution against very high concentrations of bacteria.

# Example 2

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Example 2 was conducted to measure the effects of antimicrobial solution at various concentrations on pathogenic, indicator and spoilage populations of bacteria associated with poultry. To this end, scalder water was collected from the overflow end (entrance end) of a commercial poultry scalder. The water was autoclaved to eliminate all populations of bacterial and bacterial spores to avoid interference during the study.

The autoclaved scalder water was evaluated chemically and compared to raw scalder water to ensure that the organic material demand in raw and autoclaved scalder water were similar.

The antimicrobial composition as in Example 1 was diluted with deionized water to ratios of about 1:100, 1:150, 1:200, 1:300, and 1:400 (composition to water).

Sets of test tubes were prepared as controls by adding 9 mL of autoclaved (sterilized) scalder water to sterile polystyrene test tubes. One set was prepared as controls by adding 9 mL of autoclaved scalder water to test tubes. One set was prepared by adding 9 mL of autoclaved scalder water and 1 mL of each antimicrobial solution. The control solution, as with the previous examples, comprised a cetylpyridinium chloride solution in water.

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The pathogens Salmonella typhimurium, Listeria monocytogenes, Staphylococcus aureus, the indicator Escherichia coli, and the spoilage bacteria Pseudomonas fluorescens and Shewanella putrefaciens were grown overnight in Brian Heart infusion broth at 25 °C for 24 hours. Each bacterium was exposed to each autoclaved scalder water-sanitized combination for 2 minutes to mimic scalding. After exposure period, 1 mL of the suspension was placed into 9 mL of the Brian Heart infusion broth and vortexed. One mL of this mixture was then placed into the Bactometer module well and bacterial growth was measured. The results are presented in Tables 1-7.

The antimicrobial test solution disclosed above was found effective for eliminating populations of Salmonella, Pseudomonas, and Shewanella especially when used at concentrations of 1:150 or lower with scalder water applications. Table 1 shows the effects of antimicrobial solution at various concentrations as compared with a control solution. It can be seen from Table 1 that bacterial elimination is fairly high for a solution diluted to about 1:100. Table 1 also shows the comparative effect of the test solution on Salmonella typhimurium as compared with a control solution. It can be seen in Table 1 that the test solution diluted to about 1:100 and 1:150 is very effective in reducing colony forming units.

Table 1: The effect of Test Solution various concentrations on Salmonella typhimurium

	Detection Time (hours)				
	(bacterial elimination at 24 hours)				
	Controls Test Solution				
1 to 100	5.9	23.28			
1 to 150	5.25 19.44				
1 to 200	5.35 6.89				
1 to 300	5.2 6.33				
1 to 400	5.25 5.63				

	Log <sub>10</sub> Colony Forming Units			
	Controls Test Solution			
1 to 100	4.94	0.1		
1 to 150	5.41 0.1			
1 to 200	5.34 . 4.22			
1 to 300	5.45 4.63			
1 to 400	5.41 5.13			

The effect of the antimicrobial solution on *Listeria* is shown in Table 2. It can be seen that the test solution according to the exemplary embodiment of the invention completely eliminated populations of *Listeria* and *Staphylococcus* at all concentrations, including solutions diluted with water to about 1:400. Table 2 also shows that colony forming units were substantially eliminated by the antimicrobial solution at all concentrations.

Table 2: The effect of Test Solution at various concentrations on Listeria monocytogenes

	Detection Time (hours)				
1	(bacterial elimination at 24 hours)				
	Controls Test Solution				
1 to 100	7.15	24			
1 to 150	6.25	24			
1 to 200	7.05 24				
1 to 300	7.1 24				
1 to 400	6.7 24				
	Log <sub>10</sub> Colony Forming Units				
	Controls Test Solution				
1 to 100	5.54 0				
1 to 150	5.97 0				
1 to 200	5.59 0				
	5.56 0				
1 to 300	5.56	U			

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Table 3 shows the comparative effects of various dilutions of the antimicrobial test solution on *E. coli*. As shown, the test solution was able to eliminate populations of *E. coli* at a dilution of about 1:100. At dilutions of about 1:150 (or lower) the test solution was able to eliminate all species tested with the exception of *E. coli*. Because *E. coli* is not a pathogen, it is not necessary that it be eliminated at the scalder. Instead, it can be eliminated later in the process. For this reason, a water dilution of about 1:150 has been found to be suitable for the scalder.

Table 3: The effect of Test Solution at various concentrations on Escherichia coli

	Detection Time (hours)					
	(bacterial elimination at 24 hours)					
	Controls Test Solution					
1 to 100	4.85	24				
1 to 150	4.3	5.07				
1 to 200	4.45 5.72					
1 to 300	4.5 5.03					
1 to 400	4.1 4.98					
	Log <sub>10</sub> Colony Forming Units					
	Controls Test Solution					
1 to 100	5.13 0					
1 to 150	5.67 4.92					
1 to 200	5.52 4.29					
1 to 300	5.47	4.96				
1 to 400	5.86 5					

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Table 4 shows the comparative effects of the test solution at different concentration on *Staphylococcus aureus*.

Table 4: The effect of Test Solution at various concentrations on Staphylococcus aureus

	Detection Time (hours)				
ļ	(bacterial elimination at 24 hours)				
	Controls	Test Solution			

7.8	24	
6.9	24	
7.25	24	
7.3	24	
7.1	24	
Log <sub>10</sub> Colony	Forming Units	
Controls	Test Solution	
2.56	0	
3.32	0	
3.02	0	
2.98	0	
3.15	0	
	6.9 7.25 7.3 7.1 Log <sub>10</sub> Colony Controls 2.56 3.32 3.02 2.98	

Tables 5 and 6 comparatively show the effect of the test solution at different concentrations on *Pseudomonas fluorescens* and *Shewanella putrefaciens*.

5 Table 5: The effect of Test Solution various concentrations on Pseudomonas fluorescens

	Detection Time (hours)				
	(bacterial elimination at 24 hours)				
	Controls	Controls Test Solution			
1 to 100	4.7	23.88			
1 to 150	4.1 10.29				
1 to 200	4.4 5.66				
1 to 300	4.4 4.88				
1 to 400	3.95 4.81				

Table 6: The effect of Test Solution at various concentrations on Shewanella putrefaciens

	Detection Time (hours)					
	(bacterial elimination at 24 hours)					
	Controls Test Solution					
1 to 100	6.75	24				
1 to 150	6.05 24					

1 to 200	6.65	6.89
1 to 300	6.6	11.12
1 to 400	6.2	11.61

Finally, Table 7 is a graph that comparatively shows the effect of the antimicrobial solution for eliminating colony forming units of *Campylobacter jejuni* at a dilution of 1:150. These results verify that the antimicrobial test solution disclosed herein is superior over the conventional compositions for treating microorganisms.

Table 7: The effect of Test Solution on Campylobacter jejuni at a dilution of 1:150

	Log <sub>10</sub> Colony Forming Units			
	Controls Test Solution			
1 to 150	4.6	0		

#### Example 3

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The effects of the antimicrobial test solution on pathogenic indicator and spoilage populations of bacteria associated with broiler chicken carcasses attached to food contact surfaces were studied.

The pathogens, Salmonella typhimurium, Listeria monocytogenes, Staphylococcus aureus, the indictor Escherichia coli, and the spoilage bacteria Pseudomonas Fluorescens and Shewanella putrefaciens were grown overnight in Brian Heart infusion broth at 25 °C for 24 hours. Five sterile TEFLON<sup>TM</sup> coupons were coated with 0.200 mL of each of the pathogens, the indicator or the spoilage species of bacteria (total of 30 coupons). The bacterial inocula were allowed to dry on the surface of the coupon for 4 hours. Each coupon was sprayed for 10 seconds (3 separate sprays) using a 1:100 concentration of the test solution. Each coupon was completely coated with 30 mL solution of this solution. No sanitizer residual or wet appearance occurred. After the exposure period each coupon was rinsed in 100 mL of sterile 1% buffered peptone broth. One mL of this mixture was then placed into 9 mL of Brian Heart infusion broth and then 1 mL of this mixture was placed into the Bactometer module well for measuring bacterial growth.

A control solution as disclosed above was prepared. In addition, an antimicrobial solution as disclosed herein was prepared for testing purposes. A sample of the coupons coated with the control solution and the balance was coated with the disclosed

antimicrobial solution. In both applications, electrostatic coating technique was used to adherently coat the entire surface of the coupon substrate.

The results are shown at Figure 5. It can be seen from Figure 5 that the test solution was extremely effective in eliminating populations of Salmonella, Listeria, Staphylococcus, E. coli, and Pseudomonas on food-contact surfaces. This method is effective for treating and sanitizing food-contact surfaces before or after processing operation.

# Example 4

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The effectiveness of various compositions was tested at several concentrations on *E. coli*, *Salmonella typhimurium*, and *Listeria monocytogenes*. Specifically, stock solutions were prepared from various combinations of the components cetylpyridinium chloride (component "A"), alkyl dimethyl benzyl ammonium chloride (component "B"), cetyl trimethyl ammonium chloride (component "C"), and trichloromelamine (component "D"). The various stock solutions were then diluted with de-ionized water to form 1% v/v (*i.e.*, 10,000 ppm) solutions, 0.0502% v/v (*i.e.*, 502 ppm) solutions, and 0.0015% v/v (*i.e.*, 15 ppm) solutions. A control solution of de-ionized water was also prepared. The various dilute solutions were then contacted to agar plates inoculated with *E. coli*, *Salmonella*, or *Listeria* and incubated for 48 hours at 35°C. Each test was run in triplicate. The results in terms of CFU and log<sub>10</sub> CFU are shown in Tables 8-10.

At a 1% concentration, all of the various compositions resulted in 100 % growth inhibition (i.e., 0 CFU or no growth).

Table 8: Compositions at 1% against E. coli, Salmonella, and Listeria

	F	E. Coli	Sa	Salmonella		isteria
Solution	CFU	Log	CFU	Log	CFU	Log
A & B	0	0	0	0	0	0
A & C	0	0	0	0	0	0
A & D	0	0	0	0	0	0
B & C	0	0	0	0	0	0
B & D	0	0	0	0	0	0
C & D	0	0	0	0	0	0
A & B & C	0	0	0	0	0	0
A & C & D	0	0	0	0	О	0

A & B & D	0	0	0	0	0	0
B & C & D	0	0	0	0 .	0	0
Control	6400	3.806	3600	3.556	108000	5.033
	8800	3.944	3800	3.580	11000	4.041
	13800	4.140	6400	3.806	148000	5.170
		Log Avg.		Log Avg.		Log Avg.
		3.964		3.647		4.748

Table 9: Compositions at 502 ppm against E. coli, Salmonella, and Listeria

	E. coli			Salmonella			Listeria		
Solution	CFU	Log	Log Avg.	CFU	Log	Log Avg.	CFU	Log	Log Avg.
A & B	7900	3.897	3.766	8800	3.944	3.051	2400	3.380	2.906
	1540	3.188		168	2.225		390	2.591	
	16300	4.212		960	2.982		560	2.748	
A & C	28400	4.453	4.436	1080	3.033	3.386	77	1.886	2.654
	29600	4.471		12800	4.107		1502	3.177	
	24100	4.382		1040	3.017		790	2.898	
A & D	0	0	0	8500	3.929	3.488	1240	3.093	3.105
	0	0		7600	3.881		1630	3.212	
	0	0		450	2.653		1020	3.009	
B & C	1440	3.158	3.338	0	0	0	1840	3.265	1.088
	2240	3.350		0	0		0	0	
	3200	3.505		0	0		0	0	
B & D	1820	3.260	3.327	0	0	0.055	0	0	0
	4100	3.613		0	0		0	0	
	1280	3.107		44	1.643		0	0	
C & D	20000	4.301	4.366	45	1.653	2.221	1360	3.134	3.097
	25600	4.408		310	2.491		1120	3.049	
	24400	4.387		330	2.519		1280	3.107	
A & B & C	16800	4.225	4.392	4600	3.663	3.681	440	2.643	2.978
	30000	4.477		5000	3.699		1110	3.045	

	29800	4.474		4800	3.681		1760	3.246	
A & B & D	0	0	0	8200	3.914	3.840	610	2.785	2.787
	0	0		7600	3.881		660	2.820	
	0	0		5300	3.724		570	2.756	
A & C & D	0	0	0	890	2.949	3.402	0	0	0
	0	0		1290	3.111		0	0	
	0	0		14000	4.146		0	0	
B & C & D	28800	4.459	4.427	132	2.121	2.508	1800	3.255	2.882
	24200	4.384		590	2.771	i	430	2.633	
	27400	4.438		430	2.633		570	2.756	
Control	366000	5.563	5.557	5200	3.716	3.697	360	2.556	2.908
	290000	5.462		3600	3.556		420	2.623	
	442000	5.645		6600	3.820		3500	3.544	

Table 10: Compositions at 15 ppm against E. coli, Salmonella, and Listeria

	E. coli				Salmonel	la	Listeria		
Solution	CFU	Log	Log Avg.	CFU	Log	Log Avg.	CFU	Log	Log Avg.
A & B	25600	4.121	4.090	66	1.820	1.381	12800	4.107	4.002
	24000	4.380		35	1.544		11000	4.041	
	20800	4.318		6	0.778	ļ	7200	3.857	
A & C	15200	4.182	4.100	1840	3.265	3.202	15200	4.183	4.189
	22000	4.342		1440	3.158		18000	4.255	
	18800	4.274		1520	3.182		13500	4.130	
A & D	28000	4.447	4.110	1760	3.246	3.199	11000	4.041	4.054
	27600	4.441		1480	3.170		13500	4.130	
	13600	4.134		1520	3.182		9800	3.991	
B & C	20800	4.318	4.008	420	2.623	2.852	13200	4.121	3.959
	17600	4.246		760	2.881		6800	3.833	
	24000	4.380	•	1130	3.053		8400	3.924	
B & D	18400	4.265	4.101	680	2.833	3.025	12000	4.079	4.084
	12800	4.107		1040	3.017		7600	3.881	

ļ	15200	4.182		1680	3.225		19600	4.292	
C & D	26000	4.415	4.097	1840	3.265	3.206	15700	4.196	4.082
	31200	4.494		1280	3.107		13500	4.130	
	28000	4.447		1760	3.246	i	8300	3.919	
A & B & C	12000	4.079	4.046	1920	3.283	3.159	26000	4.415	4.296
	21200	4.326		1000	3.000		24800	4.395	
•	14400	4.158		1560	3.193		12000	4.079	
A & B & D	15600	4.193	4.040	1600	3.204	3.252	0	0	0
	9600	3.982		1920	3.283		0	0	
	8800	3.945		1840	3.265		0	0	
A & C & D	12700	4.104	4.020	2200	3.342	3.313	0	0	0
	9800	3.991		2240	3.350		0	0	
	9200	3.964		1760	3.246		0	0	
B & C & D	11700	4.068	4.143	5700	3.756	3.649	0	0	0
	17800	4.250		1760	3.246		0	0	
	12900	4.111		8800	3.945		0	0	
Control	25200	4.401	4.180	800	2.903	3.137	20000	4.301	4.094
	8000	3.903		1920	3.283		10000	4.000	
	17200	4.236		1680	3.225		9600	3.982	

Although the exemplary embodiments provided herein are directed to a poultry processing line, it will be understood that the disclosed invention can be applied to other aqueous environments in general without departing from the spirit of the invention.

#### 5 Example 5

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A pool located in a Caribbean Resort containing 47,000 gallons (178 kL) water and a constant daily bather load experienced poor control of water clarity using 5 lbs. (2.3 kg) per day dry calcium hypochlorite containing 67% available chorine (approximately 12.5 ppm as dry product). A test composition as described in Examples 1-3 was added to the perimeter of the pool with the sand filtration system running continuously to provide good recirculation. An amount equivalent to 0.56 ppm dry weight basis and 7.5 ppm as liquid product was added to the bulk pool water. Within 24 hours a significant improvement in visual water quality was noted and within 48 hours the water quality was crystal clear and acceptable for use.

No further chemical additions occurred for 6 weeks in this pool, including the normal 5 lbs (2.3 kg) per day calcium hypochlorite treatment. During week 7, a once per week maintenance dosage of the test composition was added at a 0.112 ppm rate (dry basis) and 1.5 ppm as liquid product. Excellent water quality continued with no change in daily bather load.

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It will also be understood by those of skill in the art that although the components of the exemplary embodiments are represented in their respective weight percent, the ratios may nonetheless be varied to include molar or volume percent of each component.

It will be apparent to those skilled in the art that various modifications and variations can be made in the present invention without departing from the scope or spirit of the invention. Other embodiments of the invention will be apparent to those skilled in the art from consideration of the specification and practice of the invention disclosed herein. It is intended that the specification and examples be considered as exemplary only, with a true scope and spirit of the invention being indicated by the following claims.

#### **CLAIMS**

#### What is claimed is:

1. A method of treating a microorganism in an aqueous environment, comprising contacting the aqueous environment with an effective amount of an antimicrobial composition comprising any two components selected from the group consisting of an aliphatic heteroaryl salt, trichloromelamine, an aliphatic benzylalkyl ammonium salt, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt, wherein when two of the listed components are present, the other listed components are not present.

- 2. The method of claim 1, wherein the microorganism comprises the bacteria Pseudomonas aeruginosa, Enterobacter aerogenes, Proteus vulgaris, Staphylococcus aureus, Bacillus cereus, Escherichia coli, or Legionella pneumophila.
- 3. The method of claim 1, wherein the microorganism comprises the fungi

  Aspergilium niger, Aspergilius phoenicis, Penicillium funiculosum, Alternaria

  alternata, Cladosporium cladosporioides, Endomyces geotrichum, Aerobasidium

  pullulan, or Chaetomium globosum.
- 4. The method of claim 1, wherein the microorganism comprises the algae Oscillatoria geminate, Nostoc sp, Phormidium foveolarum, Chlorella vulgaris, Chlorella pyrenoidosa, Scenedesmus sp, Ulthrix subtilissima, or Tribonema aequale.
- 5. The method of claim 1, wherein the aqueous environment is a swimming pool, hot tube, or fountain.
- 6. The method of claim 1, wherein the aqueous environment is a cooling water, oil field water, mining process water, food processing water, papermaking water, sugar reprocessing water, or carpet manufacturing water.

7. The method of claim 1, wherein the aqueous environment comprises at least one of a reservoir, well, irrigation line, hose, aqua duct, sprayer, sprinkler, drip line, or soaker hose.

- 8. The method of claim 1, wherein the aqueous environment is an aqua culture water.
- 9. The method of claim 1, wherein the aqueous environment comprises a cellulosic material.
- 10. The method of claim 1, wherein the aqueous environment has a pH of from about 5 to about 9.
- 11. The method of claim 1, wherein the aqueous environment has a temperature of from about 4°C to about 45°C.
- 12. The method of claim 1, wherein the aqueous environment further comprises chloride or bromine.
- 13. The method of claim 1, wherein the aliphatic heteroaryl salt is present and comprises an alkylpyridinium halide.
- 14. The method of claim 13, wherein the alkylpyridinium halide comprises cetylpyridinium chloride, cetylpyridinium bromide, or a mixture thereof.
- 15. The method of claim 1, wherein the aliphatic benzylalkyl ammonium salt is present and comprises alkyl dimethyl benzyl ammonium chloride, alkyl methylethyl benzyl ammonium bromide, or a mixture thereof.
- 16. The method of claim 1, wherein trichloromelamine is present and the aliphatic heteroaryl salt is present and comprises cetylpyridinium chloride.

17. The method of claim 1, wherein trichloromelamine is present and the aliphatic benzylalkyl ammonium salt is present and comprises alkyl dimethyl benzyl ammonium chloride.

- 18. The method of claim 1, wherein the aliphatic benzylalkyl ammonium salt is present and comprises alkyl dimethyl benzyl ammonium chloride and the tetraalkyl ammonium is present and comprises cetyl trimethyl ammonium chloride.
- 19. The method of claim 1, wherein after contacting the aqueous environment, the composition has a concentration of from about 20 to about 600 ppm, based on the combined components.
- 20. A method of treating a microorganism in an aqueous environment, comprising contacting the aqueous environment with an effective amount of an antimicrobial composition, comprising:
  - a. an aliphatic benzylalkyl ammonium salt;
  - b. trichloromelamine; and
  - an ammonium salt selected from the group consisting of an aliphatic heteroaryl salt, a dialiphatic dialkyl ammonium salt, and a tetraalkyl ammonium salt,

wherein when the ammonium salt is the aliphatic heteroaryl salt, the composition does not contain the dialiphatic dialkyl ammonium salt or the tetraalkyl ammonium salt,

wherein when the ammonium salt is the dialiphatic dialkyl ammonium salt, the composition does not contain the aliphatic heteroaryl salt or the tetraalkyl ammonium salt, and

wherein when the ammonium salt is the tetraalkyl ammonium salt, the composition does not contain the aliphatic heteroaryl salt or the dialiphatic dialkyl ammonium salt.

21. The method of claim 20, wherein the microorganism comprises the bacteria Pseudomonas aeruginosa, Enterobacter aerogenes, Proteus vulgaris,

Staphylococcus aureus, Bacillus cereus, Escherichia coli, or Legionella pneumophila.

- 22. The method of claim 20, wherein the microorganism comprises the fungi

  Aspergilium niger, Aspergilius phoenicis, Penicillium funiculosum, Alternaria

  alternata, Cladosporium cladosporioides, Endomyces geotrichum, Aerobasidium

  pullulan, or Chaetomium globosum.
- 23. The method of claim 20, wherein the microorganism comprises the algae Oscillatoria geminate, Nostoc sp, Phormidium foveolarum, Chlorella vulgaris, Chlorella pyrenoidosa, Scenedesmus sp, Ulthrix subtilissima, or Tribonema aequale.
- 24. The method of claim 20, wherein the aqueous environment is a swimming pool, hot tube, or fountain.
- 25. The method of claim 20, wherein the aqueous environment is a cooling water, oil field water, mining process water, food processing water, papermaking water, sugar reprocessing water, or carpet manufacturing water.
- 26. The method of claim 20, wherein the aqueous environment is an aqua culture water.
- 27. The method of claim 20, wherein the aqueous environment comprises at least one of a reservoir, well, irrigation line, hose, aqua duct, sprayer, sprinkler, drip line, or soaker hose.
- 28. The method of claim 20, wherein the aqueous environment comprises a cellulosic material.
- 29. The method of claim 20, wherein the aqueous environment has a pH of from about 5 to about 9.

30. The method of claim 20, wherein the aqueous environment has a temperature of from about 4°C to about 45°C.

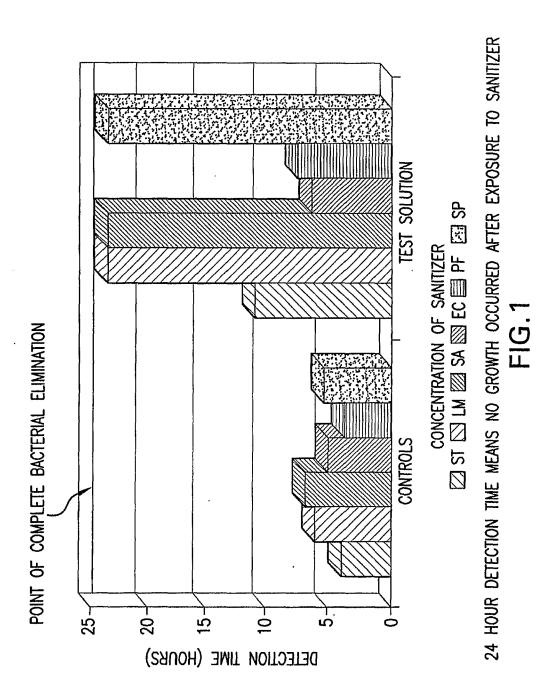
- 31. The method of claim 20, wherein the aqueous environment further comprises chloride or bromine.
- 32. The method of claim 20, wherein the aliphatic heteroaryl salt comprises an alkylpyridinium halide.
- 33. The method of claim 20, wherein the aliphatic benzylalkyl ammonium salt comprises alkyl dimethyl benzyl ammonium halide, alkyl methylethyl benzyl ammonium halide, or a mixture thereof.
- 34. The method of claim 20, wherein the ammonium salt is the dialiphatic dialkyl ammonium salt and comprises didodecyl dimethyl ammonium halide, ditetradecyl dimethyl ammonium halide, dihexadecyl dimethyl ammonium halide, or a mixture thereof.
- 35. The method of claim 20, wherein the ammonium salt is the tetraalkyl ammonium salt and comprises cetyl trimethyl ammonium halide, lauryl trimethyl ammonium halide, myristyl trimethyl ammonium halide, stearyl trimethyl ammonium halide, arachidyl trimethyl ammonium halide, or a mixture thereof.
- 36. The method of claim 20, wherein after contacting the aqueous environment, the composition has a concentration of from about 20 to about 600 ppm, based on the aliphatic benzylalkyl ammonium salt component.
- 37. A method of treating a microorganism in an aqueous environment, comprising contacting the aqueous environment with an effective amount of an antimicrobial composition, comprising:
  - a. an aliphatic benzylalkyl ammonium salt;
  - b. trichloromelamine; and
  - c. two ammonium salts selected from the group consisting of an aliphatic heteroaryl salt and a dialiphatic dialkyl ammonium salt, a aliphatic

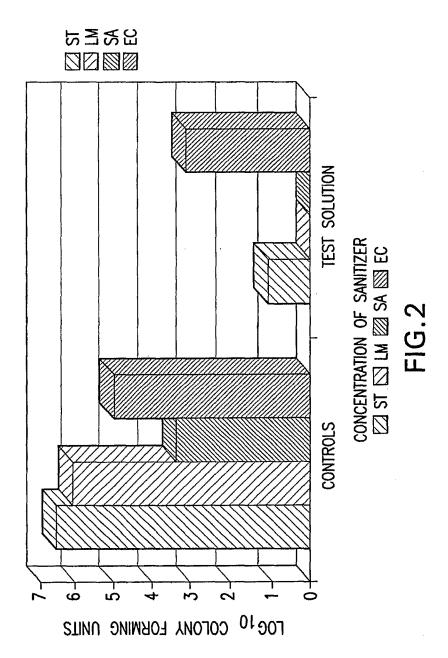
heteroaryl ammonium salt and a tetraalkyl ammonium salt, or a dialiphatic dialkyl ammonium salt and a tetraalkyl ammonium salt.

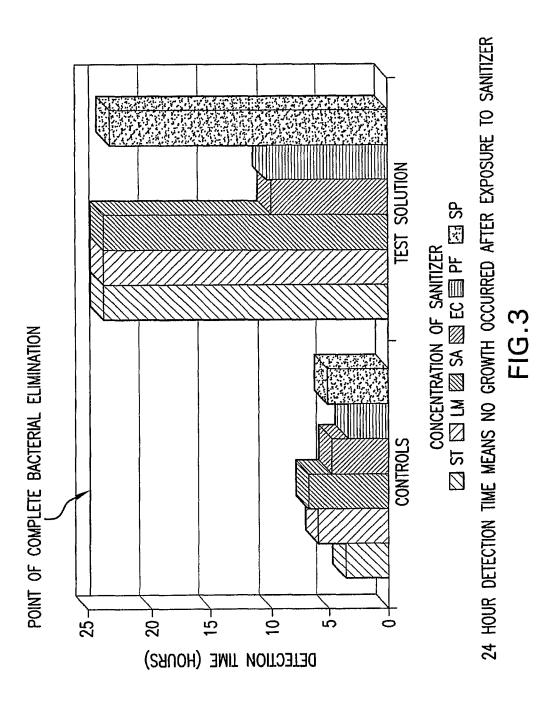
- 38. The method of claim 37, wherein the microorganism comprises the bacteria Pseudomonas aeruginosa, Enterobacter aerogenes, Proteus vulgaris, Staphylococcus aureus, Bacillus cereus, Escherichia coli, or Legionella pneumophila.
- 39. The method of claim 37, wherein the microorganism comprises the fungi Aspergilium niger, Aspergilius phoenicis, Penicillium funiculosum, Alternaria alternata, Cladosporium cladosporioides, Endomyces geotrichum, Aerobasidium pullulan, or Chaetomium globosum.
- 40. The method of claim 37, wherein the microorganism comprises the algae Oscillatoria geminate, Nostoc sp, Phormidium foveolarum, Chlorella vulgaris, Chlorella pyrenoidosa, Scenedesmus sp, Ulthrix subtilissima, or Tribonema aequale.
- 41. The method of claim 37, wherein the aqueous environment is a swimming pool, hot tube, or fountain.
- 42. The method of claim 37, wherein the aqueous environment is a cooling water, oil field water, mining process water, food processing water, papermaking water, sugar reprocessing water, or carpet manufacturing water.
- 43. The method of claim 37, wherein the aqueous environment is an aqua culture water.
- The method of claim 37, wherein the aqueous environment comprises at least one of a reservoir, well, irrigation line, hose, aqua duct, sprayer, sprinkler, drip line, or soaker hose.
- 45. The method of claim 37, wherein the aqueous environment comprises a cellulosic material.

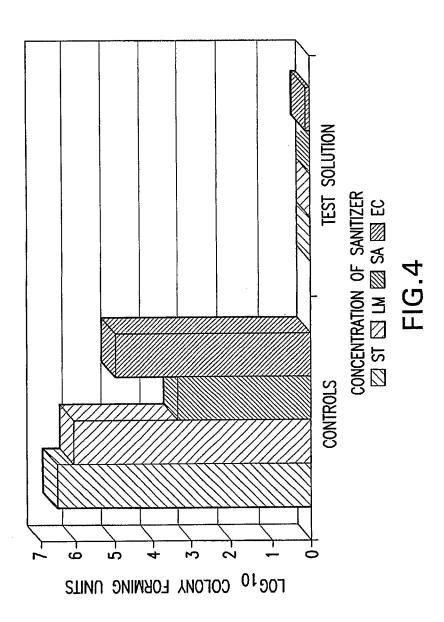
46. The method of claim 37, wherein the aqueous environment has a pH of from about 5 to about 9.

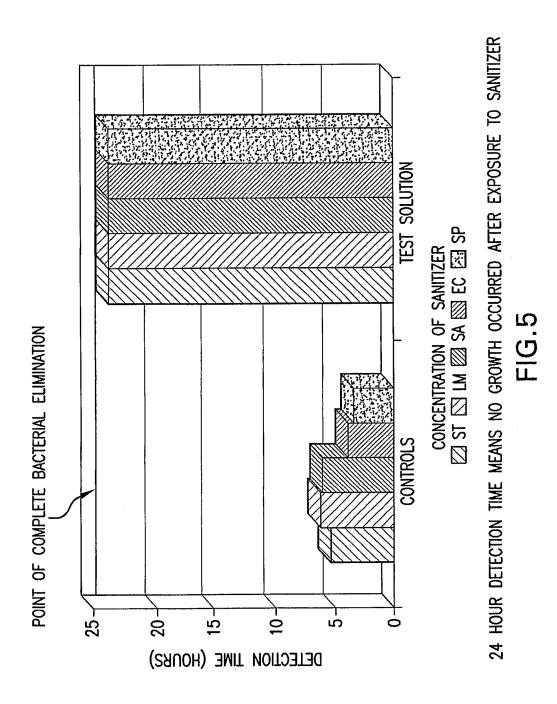
- 47. The method of claim 37, wherein the aqueous environment has a temperature of from about 4°C to about 45°C.
- 48. The method of claim 37, wherein the aqueous environment further comprises chloride or bromine.
- 49. The method of claim 37, wherein after contacting the aqueous environment, the composition has a concentration of from about 20 to about 600 ppm, based on the aliphatic heteroaryl salt component.











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